Undressing a Collective Intersubband Excitation in a Quantum Well

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We have experimentally measured the 1–2 intersubband absorption in a single 40 nm wide modulation-doped $Al_{0.3}Ga_{0.7}As/GaAs$ square quantum well as a function of frequency, intensity, and charge density. The low-intensity depolarization-shifted absorption occurs near 80 cm^{-1} (10 meV or 2.4 THz), nearly 30% higher than the intersubband spacing. At higher intensities, the absorption peak shifts to lower frequencies. Our data are in good agreement with a theory proposed by Załużny, which attributes the redshift to a reduction in the depolarization shift as the excited subband becomes populated.

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The properties of elementary excitations in solids are renormalized ("dressed") by interactions with electrons, phonons, or other particles. An example is the absorption of light by a metal; electrons in a metal are nearly free, and the lowest excited state has an energy only infinitesimally greater than the ground state. However, interactions with other electrons dress the frequency at which light is absorbed, shifting it from zero frequency (dc) to the plasma frequency.

A single electron in a semiconductor quantum well would have no electron-electron interactions. It would obey the single-particle, linear Schrödinger equation, and resonantly absorb light at a frequency equal to the difference between quantized subband energies. Many electrons are present in real quantum wells. Electron-electron interactions cause a static modification to the shape of the quantum well potential, but also allow electrons to dynamically screen oscillating fields inside the well. This screening blueshifts the frequency at which radiation is absorbed from the intersubband spacing to the dressed frequency at which collective oscillations of the entire electron gas occur [1]. This dressing of the intersubband absorption frequency by electron-electron interactions is called the *depolarization shift*.

Intersubband dynamics have been well understood in narrow quantum wells which have intersubband transitions greater than the LO phonon energy of 36 meV [2–4]. In such wells, the depolarization shift is insignificant at ordinary charge densities. In wider quantum wells, the intersubband separation is smaller, and the depolarization shift becomes a larger fraction of the absorption frequency. In this Letter, we report the first steady-state measurements of the intensity-dependent absorption line shape in a wide quantum well [5]. We find that the intersubband absorption is "undressed" by intense resonant excitation, as first predicted by Załużny: As intensity is increased, the resonant frequency moves to the red from its depolarization-shifted value toward the intersubband spacing [6].

The 40.0 nm wide GaAs square well used in our measurements was grown by molecular beam epitaxy. The potential barriers on each side of the well are 675 nm of $Al_{0.3}Ga_{0.7}As$. The well is symmetrically modulation-doped by silicon layers of sheet density 1.3×10^{12} cm^{-2} which are placed 125 nm from each side of the well. The relatively large distance between the donors and the well ensured that the charge transferred into the well was insufficient to begin occupying the second subband at low temperatures. The mobility at 4.2 K was 360 000 cm^2/V s, as measured by magnetotransport. All experiments reported here were performed on a sample 1.01 cm long, 0.7 cm wide, and 445 μm thick. An 80 nm thick aluminum gate was evaporated onto the front of the sample, with a similar layer evaporated on the back of the substrate. Ohmic contacts on the sample corners provide electrical connection to the electrons in the well. The charge density in the well was measured *in situ* by capacitance-voltage profiling.

Far-infrared radiation (FIR) from the UCSB free-electron laser was used to excite the sample. The pulses were 2.5 μs long, with peak powers of ~1 kW. The sample was mounted in a vacuum on the cold finger of a continuous-flow cryostat. All measurements were performed with the sample near 10 K. FIR was focused onto the edge of the sample, with the electric field parallel to the growth direction of the sample. A 3 mm thick plexiglass beam block prevented most of the FIR from leaking around the sample. The thick aluminum layers on the front and back serve to confine the FIR field within the sample. The transmitted FIR was measured using a 4.2 K bolometer. The entire experiment was performed in a dry nitrogen atmosphere to eliminate absorption of the FIR by water vapor. To avoid oscillations in transmittance due to standing waves within the sample, a broad-band antireflection coating was evaporated onto the edge of the sample, as described by McKnight *et al.* [7].

The large dots in Figs. 1 and 2 are attenuation coefficients for our sample, measured at gate biases of -0.5 V (sheet density $N_s = 5.7 \times 10^{10} e/\mathrm{cm}^2$) and 0 V ($N_s = 1.1 \times 10^{11} e/\mathrm{cm}^2$), respectively. The attenuation coefficient is defined as $\alpha = -\ln[T(V_G)/T(-2.0\ \mathrm{V})]/L$, where L is the length of the sample and $T(V)$ is the intensity transmitted at a gate voltage V_G. (At -2.0 V gate bias, the well was completely depleted of electrons, providing an optical reference.) The top curves show the absorption at low intensity, where it is independent of intensity. The three lower curves show the absorption line shape for intensities comparable to the saturation intensity [8]. As the intensity increases, the peak shifts to lower frequencies and becomes strongly asymmetric. The statistical uncertainty shown does not take into account systematic errors in the transmission of the sample. The shift to lower frequency and distortion of the line shape are more pronounced for Fig. 2 than for Fig. 1, as would be expected from the reduction of a larger depolarization shift, due to a larger charge density.

The experimental results were modeled using a theory by Załużny [6]. This theory calculates the steady-state nonlinear intersubband absorption of a strongly driven square quantum well including the depolarization shift. It considers two subbands in the rotating wave approximation, and uses the Hartree approximation but ignores exchange and correlation. The time evolution of the density matrix ρ is given by [6]

$$\frac{\partial \rho}{\partial t} = (1/i\hbar)[\mathcal{H}_0 + \mathcal{H}'(t), \rho] - R\rho\,, \qquad (1)$$

where the unperturbed Hamiltonian $\mathcal{H}_0$ describes the electrons and their static interactions, $\mathcal{H}'(t)$ is the perturbing Hamiltonian, and R is the relaxation operator. The perturbation Hamiltonian is the sum of the energy of the electrons in the field, plus the correction to the static Hartree energy due to the changes in the electron distribution caused by the oscillating field,

$$\mathcal{H}'(t) = eD(t)z - (4\pi e^2/\epsilon)\int_{-\infty}^{z} dz' \int_{-\infty}^{z'} dz'' \Delta n(z'', t)\,, \qquad (2)$$

where $D(t)$ is the external oscillating electric field, $\Delta n(z,t)$ is the change in the electron distribution due to the oscillating field, and ϵ is the background dielectric constant of the semiconductor.

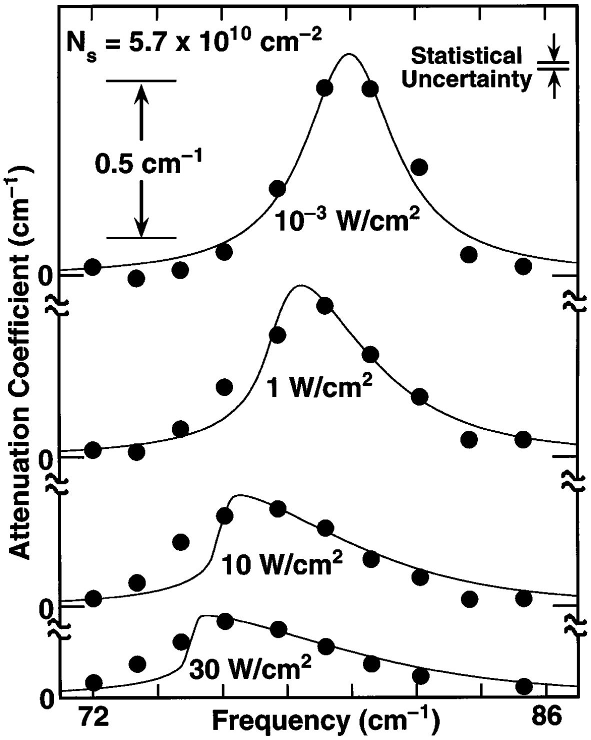

FIG. 1. FIR absorption vs frequency at various intensities for $V_G = -0.5$ V, with $N_s = 5.7 \times 10^{10}$ cm^{-2}. The large dots are experimental data, and the curves are calculated from a theory by Załużny. For 1 W/cm^2, $\tau_1 = 760$ ps and $I_{\mathrm{sat}} = 2.5$ W/cm^2. For 10 W/cm^2, $\tau_1 = 230$ ps and $I_{\mathrm{sat}} = 8.3$ W/cm^2. For 30 W/cm^2, $\tau_1 = 130$ ps and $I_{\mathrm{sat}} = 15$ W/cm^2.

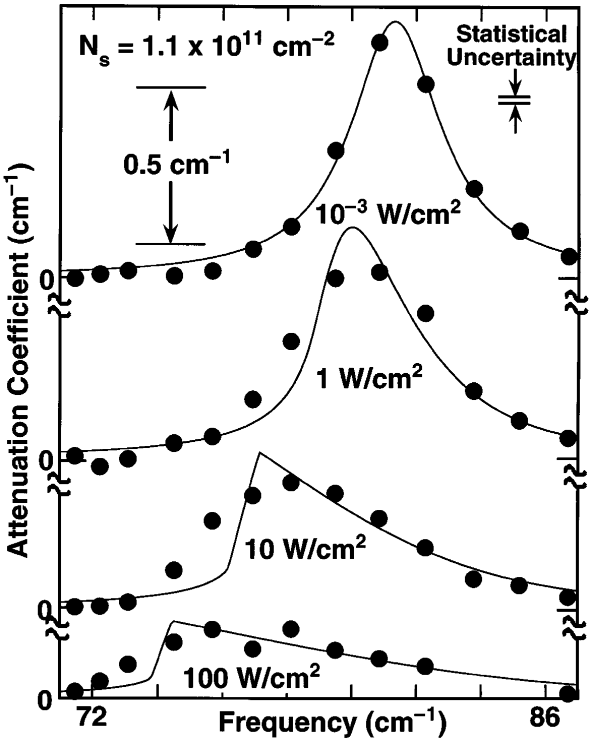

FIG. 2. FIR absorption vs frequency at various intensities for $V_G = 0$ V, with $N_s = 1.1 \times 10^{11}$ cm^{-2}. The large dots are experimental data, and the solid curves are calculated from a theory by Załużny. For 1 W/cm^2, $\tau_1 = 530$ ps and $I_{\mathrm{sat}} = 6.5$ W/cm^2. For 10 W/cm^2, $\tau_1 = 240$ ps and $I_{\mathrm{sat}} = 14.5$ W/cm^2. For 100 W/cm^2, $\tau_1 = 70$ ps and $I_{\mathrm{sat}} = 50$ W/cm^2.

Załużny's theory includes in a nonperturbative manner the corrections to the Hartree potential caused by the redistribution of electrons in the applied oscillating field. These redistributions are responsible for dynamic screening and the depolarization shift. Załużny's theory is able to correctly calculate the absorption of the collective intersubband excitation even when the second subband is heavily populated by FIR excitation. (Changes to the static self-consistent potential due to the redistribution of charge in real space are not included, but should not be an important effect in our sample.)

The input parameters of the theory (see Table I) are the dressed intersubband spacing $\hbar\tilde{\omega}_{21}$, the bare intersubband spacing $\hbar\omega_{21}$, the matrix element $z_{21} \equiv \langle\chi_2|z|\chi_1\rangle$ where $|\chi_n\rangle$ are the self-consistent wave functions that solve the time-independent Schrödinger equation in the Hartree approximation, the energy and momentum relaxation rates Γ_1 and Γ_2 (assumed by Załużny to be independent of the intensity), and the FIR intensity I. The theory by Załużny predicts a Lorentzian line shape for the absorption at low intensities. We used the parameters from a Lorentzian fit of the low-intensity results to obtain the dressed intersubband absorption frequency $\tilde{\omega}_{21}$ and the elastic relaxation rate Γ_2. We have assumed a homogeneously broadened line. The bare intersubband spacing ($\hbar\omega_{21}$) was computed in the local density approximation for density functional theory following the procedure developed by Ando, Fowler, and Stern [1]. A well width of 40.2 nm gave the best agreement with experimentally measured absorption frequencies and is within an atomic monolayer of the design width of 40.0 nm. These simulations included exchange-correlation effects [9], as well as a dc electric field (tilt) across the well. Neither of these effects are included in the dynamic theory by Załużny. Because of uncertainties in the actual well width, charge density, and dc electric field at the well, estimates of $\hbar\omega_{21}$ are accurate within roughly ± 3 cm^{-1}.

The matrix element z_{21} can be obtained directly from experiment and from our static self-consistent simulations. The z_{21} values in Table I were derived from the experimentally measured absorption at $\omega = \tilde{\omega}_{21}$ using the relation (in CGS units) $\alpha = 4\pi\omega e^2 z_{21}^2 N_s / c\sqrt{\epsilon}\, a\hbar\Gamma_2$, where e is the electronic charge, c is the speed of light, $\epsilon = 13$ is the dc dielectric constant, N_s is the electronic sheet density, and a is the thickness of the wafer [10]. The simulations for $V_G = -0.5$ V yield $z_{21} = 7.6$ nm, 8% greater than the experimentally obtained value. Surprisingly, the simulations for $V_G = 0.0$ V yield a $z_{21} = 8.4$ nm which is 56% greater than the experimentally deduced value. We attribute this discrepancy to interactions of the densely populated quantum well with the waveguide modes in the sample [11]. We presently have no detailed understanding of this effect, and it is not included in our simulations.

TABLE I. Parameters used to fit the data using the theory by Załużny: from experiment,$^+$ from simulations.*

	$V_G = -0.5$ V	$V_G = 0.0$ V
$\tilde{\omega}_{21}/2\pi^+$	79.9 cm^{-1}	81.3 cm^{-1}
$\omega_{21}/2\pi^*$	72.6 cm^{-1}	60.6 cm^{-1}
z_{21}^+	7.04 nm	5.38 nm
Γ_2^+	1.7 cm^{-1}	1.7 cm^{-1}
dc tilt*	0.31 V/μm	-0.10 V/μm

The solid curves in Figs. 1 and 2 were computed using the theory of Załużny [6] with the parameters from Table I and the experimentally measured intensities as inputs. In a region of predicted optical bistability (see below), we calculated the solution which corresponded to the system starting in an unexcited state [12]. The reduction in intensity of the beam due to the strong absorption and long path length inside the sample was included in the calculations. For each of the lower three curves, the sole fitting parameter was the energy relaxation rate $\Gamma_1 = \hbar/\tau_1$. The intensity-dependent line shape is reasonably well reproduced by the calculated curves.

Qualitatively, the absorption results may be understood as follows: Theory predicts the depolarization shift to simply be a function of $N_1 - N_2$, the difference in populations of the subbands [12]. Increasing the FIR intensity causes the population in the excited subband to increase. This reduces the depolarization shift, which causes the absorption peak to shift to lower frequencies. If the frequency of intense FIR excitation is below the low-intensity absorption frequency, then the absorption will shift down into the laser line, increasing the absorption. But if the excitation frequency is above the low-intensity resonance, the absorption of the excited system will shift down, away from the laser line, reducing the absorption. This dynamical redistribution of absorption in frequency accounts for the unusual asymmetric line shape.

Optical bistability is another prediction of the theory. At high intensities, the solid curves in Figs. 1 and 2 exhibit a sharp drop in absorption on the low-frequency side of the absorption peak. In this parameter region, the absorption can switch suddenly between two stable solutions. For the two solutions, the excited subband population is high (low) and the absorption frequency is (is not) significantly redshifted from its equilibrium value. The experimental data in Figs. 1 and 2 do not exhibit the sharp edges predicted by theory. We are currently investigating this discrepancy, which may be caused by an increase in the linewidth of the absorption during strong excitation (see below).

The energy relaxation times used for the calculated curves in Figs. 1 and 2 decrease from 760 to 130 ps and 530 to 70 ps for the low- and high-density data over the range of intensities studied here. The geometric factors which lead to a factor of 3 uncertainty in the intensity (and hence the overall magnitude of the energy relaxation time τ_1) are separately fixed for the data of Figs. 1 and 2, and do not affect relative relaxation rates. The decrease of

τ_1 with increasing intensity may occur because, at higher intensities, the electron gas is hotter, and there will be more electrons with sufficient energy to emit LO phonons. Note that the theory by Załużny includes only a constant energy relaxation time, which we have varied as a function of intensity to account for the effects of LO phonon emission by hot electrons. Our energy relaxation times, including the maximum factor of 3 we estimate from systematic errors, are similar to recently reported times [13–16]. We have estimated the relaxation time due to acoustic phonon emission [17] to be roughly 750 ps for this structure, consistent with our low-intensity results [18].

Although agreement between the theory by Załużny and our experimental results is reasonably good, a more consistent and complete theory is desirable. Such a theory would account for the variation of τ_1 with intensity. In addition, the theory would include the effects of changes in the linewidth of the absorption. If strong excitation heats the electrons above the LO phonon energy, then the fast LO phonon dephasing of the electrons will cause the linewidth of the absorption to broaden. This broadening will change the nonlinear absorption of the system and complicate the interpretation of the relaxation times and saturation intensities obtained from the experiments [19]. Finally, the theory would also incorporate the effects of exchange correlation and dc tilt, which were necessary to fit the experimentally measured $\tilde{\omega}_{21}$, but are not included in Załużny's theory.

We have shown experimentally that, as the intersubband resonance is strongly excited with resonant FIR, the absorption frequency decreases. We attribute this shift to a modification in the dynamic screening caused by the change in population of the subbands. This novel mechanism for nonlinearity is quite strong in wide quantum wells, providing a new arena for the study of interacting electrons far from equilibrium, as well as the possibility of engineering novel quasioptical devices.

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[18] However, it is possible that τ_1 increases further at intensities lower than 1 W/cm^2.

[19] Note that the uncertainties for the relaxation times quoted above do not include intensity-dependent line broadening. The broadening would only be important at high intensities, where our calculations based on the Załużny theory would overestimate the relaxation time, perhaps by as much as a factor of 2 or 3. Thus τ_1 may decrease even faster with increasing intensity than we have reported here.

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