Ultrafast Dynamic Control of Spin and Charge Density Oscillations in a GaAs Quantum Well

J. M. Bao, L. N. Pfeiffer, K. W. West, and R. Merlin

1Focus Center and Department of Physics, The University of Michigan, Ann Arbor, Michigan 48109-1120, USA
2Bell Laboratories, Lucent Technologies, Murray Hill, New Jersey 07974, USA

(Received 6 October 2003; published 8 June 2004)

We use subpicosecond laser pulses to generate and monitor in real time collective oscillations of electrons in a modulation-doped GaAs quantum well. The observed frequencies match those of intersubband spin- and charge-density excitations. Light couples to coherent density fluctuations through resonant stimulated Raman scattering. Because the spin- and charge-related modes obey different selection rules and resonant behavior, the amplitudes of the corresponding oscillations can be independently controlled by using shaped pulses of the proper polarizations.

Spin currents are the most common source of magnetism and, since spin cannot be described in classical terms, it is apparent that the majority of magnetic phenomena are ultimately a manifestation of quantum behavior. While this fact has been known for a very long time, it is only recently that methods to generate spin-polarized currents have attracted much attention, driven mainly by the possibility that novel quantum effects and devices may be uncovered [1]. Next to electrical methods, the generation of magnetic currents by optical injection has now become a very active area of research. Here III-V semiconductor quantum wells (QWs), particularly those belonging to the AlGaAs/GaAs system, play an important role owing to the spin-polarized nature of their valence band [2]. We note that these heterostructures have been used for many years to produce, by means of photoexcitation, incoherent spin-polarized electron sources for applications in nuclear and high energy physics [3]. More recently, injection of a pure spin current has been achieved in a GaAs QW through interference of one- and two-photon absorption processes [4].

In this work we use ultrafast light pulses to induce coherent density oscillations associated, separately, with the spin and charge degrees of freedom of a quasi-two-dimensional electron gas (2DEG) contained in a single GaAs QW. Studies of the ultrafast dynamics of low-lying levels of a QW have been previously reported [5–14]. Our results distinguish themselves from those studies in that we are able to differentiate collective (many-particle) from single-particle behavior and observe many-electron dynamics in real time (for recent theoretical work on intersubband excitations, see [15,16]). The method we use to generate and control the spin and charge oscillations is stimulated Raman scattering (RS) by intersubband excitations. Given that spontaneous RS is one of the main tools for probing 2DEG properties and, in particular, the quantum Hall effects [17,18], our results hold promise for elucidating the coherent dynamics of these and other 2DEG phenomena.

Our sample, grown by molecular beam epitaxy on a (001) GaAs substrate, is a 400 Å one-sided modulation-doped GaAs single QW sandwiched between Al0.3Ga0.7As barriers; see Fig. 1(a). The 2DEG originates from electrons initially bound to those Si donors in the barriers which are closest to and migrate to the QW. To reduce ionized impurity scattering and thereby enhance the mobility, these donor atoms are separated from the 2DEG by a 103 Å-thick undoped spacer [20]. A schematic energy level diagram is shown in Fig. 1(b). The excitations pertinent to our work are intersubband transitions associated with the lowest-lying states of the QW. We used transport measurements at 4.2 K to determine the sample mobility $\mu = 2.9 \times 10^6$ cm$^2$/Vs and the 2DEG areal density $n_0 = 1.9 \times 10^{11}$ cm$^{-2}$ for which the corresponding Fermi energy is $E_F = 7$ meV. The latter value is consistent with the width of the main photoluminescence (PL) feature in Fig. 1(c) [21]. From the PL data, we also get ~1.512 eV for the renormalized QW band gap [22]. Figure 1(d) shows schematically the long-wavelength limit of the excitation spectrum involving the two lowest-lying subbands. The wave vector $q$ is perpendicular to the growth axis [001]. The spectrum consists of the single-particle (SP$_{01}$) continuum delimited by $E_{01} \pm \hbar q k_F / m$ and, at small wave vectors, the collective spin-density (SD$_{01}$) and charge-density (CD$_{01}$) resonances [23]. Here, $k_F$ is the Fermi wave vector, $m$ is the electron effective mass, and $E_{01} = (E_1 - E_0)$. As shown in Fig. 1(e), Raman data of our QW at $q = 0$ (vertical transitions) reveal the expected three distinct features at 2.60 (SD$_{01}$), 2.85 (SP$_{01}$) and 3.31 (CD$_{01}$) THz. These results are typical of high-mobility samples [24–26]. Here, $z$ denotes the axis normal to the layers and $k'_y$ ($\gamma'$) is along the [110] ([110]) direction [19]. We notice that the charge (spin) related peak appears in polarized (depolarized) spectra, i.e., when the incident and scattered polarizations are parallel (orthogonal) to each other. This reflects the fact that the charge (spin) density mode transforms like the symmetric $A_1$ (antisymmetric $A_2$) representation of the $D_{2d}$ point group of the QW [24].
Time domain pump-probe experiments were performed at ~7 K in the reflection geometry using a modelocked Ti:sapphire laser which provided ~50–65 fs pulses at a repetition rate of 82 MHz. The stronger pump pulse generates coherent oscillations in the electron density, which modify the 2DEG optical constants and, thereby, perturb the weaker probe pulse which follows behind. The laser beams, of average power 5–20 mW (pump) and 2.5 mW (probe), penetrated the crystal along the z axis (hence, q = 0) and were focused onto a 300-μm-diameter spot. The pump beam was either circularly or linearly polarized, along x', while the incident probe beam was linearly polarized, along y'. We measured the pump-induced change in the field of the reflected probe beam, δE_R, as a function of the time delay between the two pulses. To obtain δE_R, also known as the coherent scattered field, we determined separately the pump-induced shift of the polarization angle of the reflected probe field, δθ, and the differential reflectivity δR ≈ |E_R + δE_R|^2 − |E_R|^2 = 2E_R · δE_R (E_R is the reflected probe field when the pump is turned off), δθ. After removal of the slowly decaying electronic background, we used linear prediction methods [28] to determine the number of oscillators and their parameters. This procedure gives three modes and fits such as those of Fig. 2 which reproduce quite accurately the experimental traces. The frequencies of the two lowest modes agree extremely well with those of SD01 and CD01 from the Raman spectra in Fig. 1(e) and, on this basis, we assign them as coherent spin- and charge-density oscillations. The weaker single-particle peak was only vaguely distinguished in the time-domain data. Our experiments show that the SD01 mode can be excited only if the pump beam is circularly polarized [Fig. 2(a), bottom trace], whereas the CD01 amplitude is largest for linearly polarized pump pulses. This selectivity, as well as the fact that the amplitude of the oscillations depends strongly on the central energy of the pulses (see below), opens the road for coherent control studies. SD01 and CD01 behave also quite differently vis-à-vis the probe detection scheme. While the CD01 contribution dominates the modulated intensity [Fig. 2(a), top trace], SD01 leads mainly to a rotation of the probe polarization. Hence, δE_R is mostly perpendicular (parallel) to the incident beam for spin (charge) oscillations. The appearance of the dominant charge-density modes in the bottom trace of Fig. 2(a) and the depolarized Raman spectrum of Fig. 1(e) is attributed to a polarization leakage (notice that, relative to CD01, the time-domain signal for SD01 is significantly smaller than the Raman signal). The remaining feature at 3.97 THz, labeled CD12, is ascribed to charge-density transitions of photoexcited electrons involving the states of energies E1 and E2 [29]. We believe that the presence of these carriers,
The following single-particle analysis provides a simple physical picture of, both, the screening behavior...
of the two types of collective modes and the associated coherent states created by the laser pulses. Following an impulsive excitation with $q = 0$, the wave function of an electron initially in the state $|k,s,0\rangle$ of the lowest subband becomes, to lowest order in the pump electric field, 
$$\Psi = |k,s,0\rangle \exp(-iE_{0t}/\hbar) + \sum_{n>0} \alpha_{ns}|k,s,n\rangle \exp(-iE_{nt}/\hbar),$$
where $\alpha_{ns}$ are constants proportional to the intensity of the pulses ($|\alpha_{ns}| \ll 1$). Hence, the quasi-2DEG density for a given spin polarization varies as
$$\delta \sigma_{\alpha}(r,t) \equiv \langle \delta \sigma_{q=0,s}\rangle = \sum_{i<k_F} \alpha_{ns}|i(k,s,n)/(k,s,0)|r \exp(-i(E_{s}-E_{n})t/\hbar) + \text{c.c.} \quad (3)$$

From (2), we have that $\alpha_{ns} = \pm \alpha_{ns}$, where the plus (minus) sign is for charge (spin) excitations. Thus, the effect of an optical pulse is to create coherent density oscillations for which the spin-up and spin-down components are either in phase (charge-density mode) or 180° out of phase (spin-density mode). Given that the two contributions add up for charge excitations (i.e., $\delta \sigma_+ = \delta \sigma_-$), these plasmonlike modes experience a restoring field whereas spin excitations remain unscreened since the corresponding motion does not change the net density $(\delta \sigma_++\delta \sigma_- = 0)$. From (1) and the expression for the dielectric tensor $\varepsilon_{ij} = \langle \partial^2 H/\partial E_i E_j \rangle$, it follows that the coherent oscillations lead to a time-varying modulation of the optical constants and, because of the symmetry properties of $C_{ij}$ and $S_{ij}$, that $\delta \mathbf{E}_R$ and the reflected probe beam must be along the same direction for charge, but perpendicular to each other for spin excitations. These selection rules are consistent with the experimental observations.

Acknowledgement is made to the donors of The Petroleum Research Fund, administered by the ACS, for partial support of this research. Work also supported by the NSF Focus Physics Frontier Center.

[19] We use the standard Porto notation: $a(b,c)d$ indicates that the incident (scattered) light propagates along the $a$ ($d$) axis, and that its polarization vector is along $b$ ($c$).