Valley Susceptibility of an Interacting Two-Dimensional Electron System

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We report direct measurements of the valley susceptibility, the change of valley population in response to an applied symmetry-breaking strain, in an AlAs two-dimensional electron system. As the two-dimensional density is reduced, the valley susceptibility dramatically increases relative to its band value, reflecting the system's strong electron-electron interaction. The increase has a remarkable resemblance to the enhancement of the spin susceptibility and establishes the analogy between the spin and valley degrees of freedom.

Currently, there is considerable interest in controlled manipulation of electron spin in semiconductors. This interest partly stems from the technological potential of spintronics, namely, the use of carrier spin to realize novel electronic devices. More important, successful manipulation of spins could also impact the more exotic field of quantum computing since many of the current proposals envision spin as the quantum bit (qubit) of information [1–3]. Here we describe measurements of another property of electrons, namely, their valley degree of freedom, in a semiconductor where they occupy multiple conduction band minima (valleys) [Fig. 1(a)]. Specifically, for a two-valley, two-dimensional electron system (2DES) in an AlAs quantum well, we have determined the "valley susceptibility", χ_v, i.e., how the valley populations respond to the application of symmetry-breaking strain. This is directly analogous to the spin susceptibility, χ_s, which specifies how the spin populations respond to an applied magnetic field [Figs. 1(d)]. Our data show that χ_v and χ_s have strikingly similar behaviors, including an interaction-induced enhancement at low electron densities. The results establish the general analogy between the spin and valley degrees of freedom, implying the potential use of valleys in applications such as quantum computing. We also discuss the implications of our results for the controversial metal-insulator transition problem in 2D carrier systems.

It is instructive to describe at the outset the expressions for the *band* values of spin and valley susceptibilities $\chi_{s,b}$ and $\chi_{v,b}$ [4]. The spin susceptibility is defined as $\chi_{s,b} = d\Delta n/dB = g_b\mu_B\rho/2$, where Δn is the net spin imbalance, B is the applied magnetic field, g_b is the band Landé g factor, and ρ is the density of states at the Fermi level. Inserting the expression $\rho = m_b/\pi\hbar^2$ for 2D electrons, we have $\chi_{s,b} = (\mu_B/2\pi\hbar^2)g_bm_b$, where m_b is the band effective mass. In analogy to spin, we can define valley susceptibility as $\chi_{v,b} = d\Delta n/d\epsilon = \rho E_{2,b} = (1/\pi\hbar^2)m_bE_{2,b}$, where Δn is the difference between the populations of the majority and minority valleys, ϵ is strain, and $E_{2,b}$ is the conduction band deformation potential [5]. In a Fermi liquid picture, the interparticle interaction results in replacement of the parameters m_b, g_b, and $E_{2,b}$ [6] by their

renormalized values m^*, g^*, and E_2^*. Note that $\chi_s \propto m^*g^*$ and $\chi_v \propto m^*E_2^*$.

Our experiments were performed on a high-mobility 2DES confined to an 11 nm-thick, modulation-doped layer of AlAs grown by molecular beam epitaxy on a (001) GaAs substrate [7]. We studied three samples, each patterned in a standard Hall bar mesa aligned with the [100] crystal direction [Figs. 1(b) and 1(c)]. Using a metal gate deposited on the sample's surface we varied the 2DES density n, between 2.5 and 9.5×10^{11} cm^{-2}. The magnetoresistance measurements were mostly performed in a liquid ^{3}He system with a base temperature of 0.3 K; data taken at $\simeq 0.03$ K in a dilution refrigerator showed no measurable dependence of χ_v on temperature.

In bulk AlAs electrons occupy three (six half) ellipsoidal conduction valleys at the six equivalent X points of the Brillouin zone [Fig. 1(a)]. For the 11 nm-wide quantum well used in our experiments, only the valleys with their major axes along [100] and [010] are occupied [7]; we refer to these as the X and Y valleys, respectively. The application of symmetry-breaking strain along [100] and [010] splits the energies of the X and Y valleys, transferring charge from one valley to the other [Fig. 1(b)]. To apply tunable strain we glued the sample to one side of a piezo-electric (piezo) stack actuator [8]. The piezo's polling direction is aligned along [100] as shown in Fig. 1(c). When bias V_P is applied to the piezo stack, it expands (shrinks) along [100] for $V_P > 0$ ($V_P < 0$) and shrinks (expands) in the [010] direction. We have confirmed that this deformation is fully transmitted to the sample and, using metal strain gauges glued on the opposite side of the piezo [Fig. 1(c)], have measured its magnitude in both [100] and [010] directions [8,9].

We determined χ_v via different techniques. The first involves measuring, at $B = 0$, sample's resistance (R_{xx}), along [100] as we induce tensile (compressive) strain and the electrons are transferred to the Y (X) valley [Fig. 1(b)]. Because of the smaller (larger) effective mass of electrons in this valley along [100], R_{xx} decreases (increases) as a function of V_P and saturates once all the electrons are transferred to the Y (X) valley [10]. We can use the onset
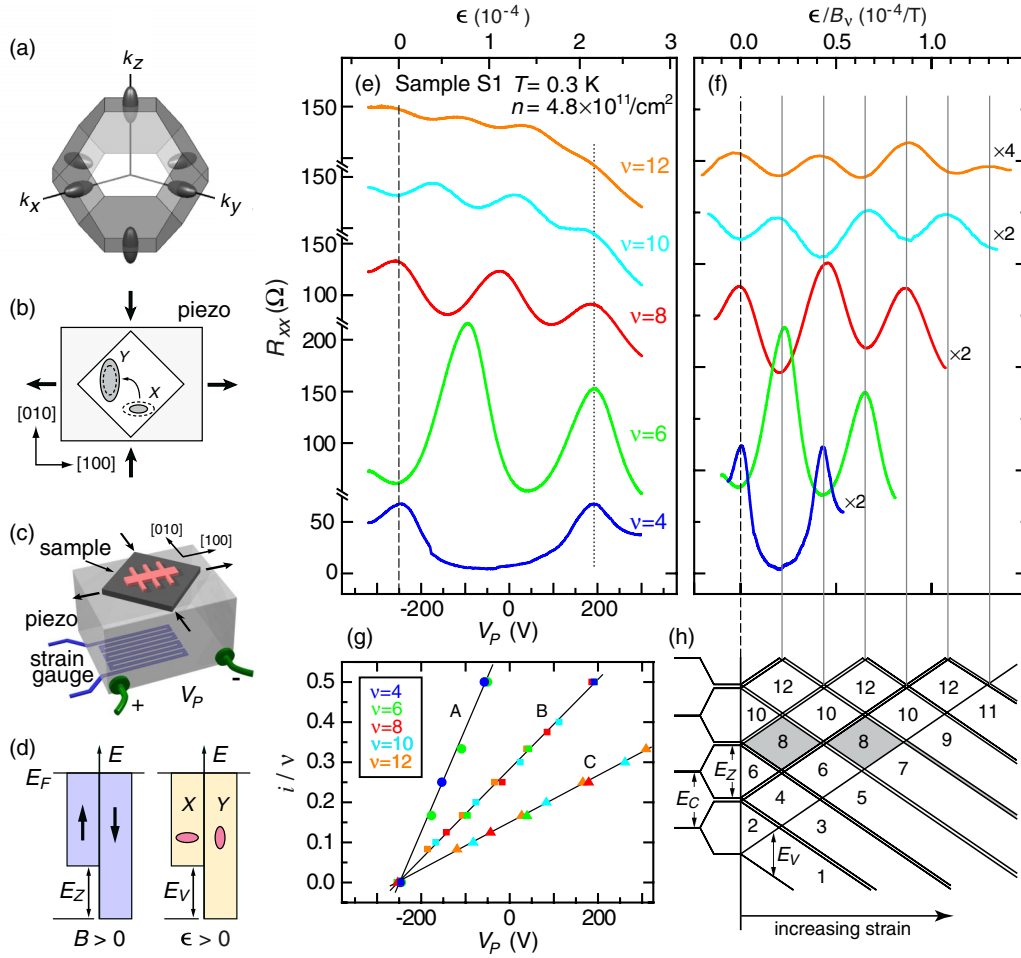
FIG. 1 (color online).　(a) The first Brillouin zone and conduction band constant energy surfaces for bulk AlAs. (b) Schematic diagram showing that electrons are transferred from the X to the Y valley upon the application of strain while the total 2DES density is independent of strain. (c) Experimental setup for the valley susceptibility measurements. (d) Energy diagram showing the spin (E_Z) and valley (E_V) subband splittings with applied magnetic field or strain, respectively. (e) Sample resistance, R_{xx}, vs the piezo bias, V_P at various filling factors ν. The traces are offset for clarity. The dashed and dotted lines, respectively, mark V_P at which X and Y are equally populated, and V_P beyond which all the electrons are transferred to the Y valley. The top axis shows the strain as measured by the strain gauge. (f) Same data as in (e) but now plotted vs a normalized horizontal (top) axis ϵ/B_ν, showing that the oscillations have the same period. The traces are offset for clarity and, for $\nu = 8$, 10, and 12, a smooth, parabolic background is subtracted from the traces to highlight the oscillations. (g) Plot of the coincidence index, i normalized by ν, showing linearity with V_P. From the slopes of the lines fitted through the data points we obtain χ_ν, and the $i = 0$ intercepts give the value of V_P at which the X and Y valleys are equally populated ($\epsilon = 0$). Three data sets A, B, and C correspond to densities of: 2.8, 4.8, and 7.6×10^{11} cm^{-2}, respectively. (h) Schematic energy fan diagram showing the relevant energies: the cyclotron (E_C), Zeeman (E_Z) and valley splitting (E_V).

of the saturation as a signature of the full electron transfer and, from the corresponding ϵ, determine χ_ν [11]. Our second technique is based on measuring the valley densities at a fixed V_P from the Shubnikov–de Haas oscillations of R_{xx} as a function of perpendicular magnetic field, $B_\perp$. The frequencies of these oscillations are proportional to the densities of the two valleys and provide a direct measure of χ_ν [12]. We quote data obtained from these two techniques later in the Letter [13]; here we would like to focus on a powerful variation of the second technique which also directly measures χ_ν.

The technique is based on the "coincidence" of the 2DES's energy levels at the Fermi energy (E_F) and entails

monitoring oscillations of R_{xx} as a function of ϵ at a *fixed* $B_\perp$ [14]. Examples of such data are shown in Fig. 1(e) for $n = 4.8 \times 10^{11}$ cm^{-2} and various values of $B_\perp$ corresponding to even Landau level (LL) filling factors $\nu = 4, 6, \ldots, 12$. Clear oscillations of R_{xx} as a function of V_P are seen. These oscillations come about because the strain-induced valley-splitting causes pairs of quantized energy levels of the 2DES in $B_\perp$ to cross at E_F. As schematically shown in the fan diagram of Fig. 1(h), there are three main energies in our system. The field $B_\perp$ quantizes the allowed energies into a set of LLs, separated by the cyclotron energy, $E_C = \hbar e B_\perp / m^*$. Because of the electron spin, each LL is further split into two levels, separated by the

Zeeman energy, $E_Z = \mu_B g^* B$. In the absence of in-plane strain, each of these energy levels in our 2DES should be twofold degenerate. By straining the sample, we remove this degeneracy and introduce a third energy, the valley-splitting, $E_V = \epsilon E_2^*$, which increases linearly with strain, as illustrated in Fig. 1(h). When an integer number of the quantized energy levels of a 2DES are exactly filled, E_F falls in an energy gap separating adjacent levels (see, e.g., the shaded area of Fig. 1(h) for the case of $\nu = 8$) and, at low temperatures, R_{xx} exhibits a local minimum. As is evident in Fig. 1(h), however, at certain values of ϵ, the energy levels corresponding to different valley- and spin-split LLs coincide at E_F. At such coincidences, the R_{xx} minimum becomes weaker or disappears altogether [14]. From the fan diagram of Fig. 1(h) we expect the weakening or strengthening of the R_{xx} minimum to be a periodic function of ϵ, or V_P, since it happens whenever E_V is an even multiple of E_C. Therefore, we can directly measure E_V (in units of E_C) and determine the valley susceptibility: $\chi_\nu = 4eB_\nu/h\Delta\epsilon$, where $\Delta\epsilon$ is the period of the R_{xx} oscillations and B_ν is $B_\perp$ of the filling factor ν at which the oscillations are measured.

Before we discuss the measured values of χ_ν as a function of density, we point out several noteworthy features of data from Figs. 1(e)–1(g). First, because of finite residual stress during the cooling of the sample and the piezo, we need a finite, cooldown-dependent V_P to attain the zero-strain condition in our experiments; this is about -250 V for the experiments of Figs. 1(e)–1(g) and is marked by a dashed vertical line in Fig. 1(e). The data of Fig. 1(e) themselves allow us to determine the zero-strain condition: at $V_P = -250$ V, R_{xx} at $\nu = 4, 8$, and 12 is at a (local) maximum while at $\nu = 6$ and 10 it is at a (local) minimum. This behavior is consistent with the energy diagram of Fig. 1(h) when no strain is present. Second, the period of the R_{xx} oscillations as a function of ϵ is larger at smaller ν (larger $B_\perp$). In Fig. 1(f) we plot R_{xx} as a function of ϵ normalized by B_ν. It is clear in this plot that the oscillations are in-phase at $\nu = 4, 8$, and 12, and that these are 180° out-of-phase with respect to the oscillations at $\nu = 6$ and 10. This is consistent with the simple fan diagram of Fig. 1(h) [15]. To quantify this periodicity and also compare the data for different densities, in Fig. 1(g) we show plots of the coincidence "index", $i = E_V/E_C$, divided by ν, vs V_P; the indices are obtained from the positions of maxima and minima of the R_{xx} oscillations and the index at the zero-strain condition is assigned to be zero. The plots in Fig. 1(g) show that the valley polarization is linear in ϵ. This linearity means that χ_ν is independent of valley polarization or ν (for $\nu \geq 4$) within our experimental uncertainty [16]. Third, in Fig. 1(e) R_{xx} at all ν shows a maximum at a particular value of ϵ (dotted vertical line at $V_P \simeq 200$ V). This maximum marks the last coincidence of the energy levels. Beyond this value of ϵ, the minority valley is completely depopulated and there are no further oscillations. We have confirmed this statement at lower

sample densities where our available range of V_P allows us to go well beyond the valley depopulation ϵ.

Figure 2 summarizes our main result, namely, the measured χ_ν as a function of density. χ_ν is enhanced at all accessible densities and particularly at lower n. This enhancement is in sharp contrast to the noninteracting picture where χ_ν should maintain its band value at all electron densities, and betrays the strong influence of electron-electron interaction in this system.

A comparison of the measured χ_ν with the *spin* susceptibility for similar samples [18] supports this conjecture. As seen in Fig. 2, χ_s and χ_ν show a remarkably similar enhancement. The increase of χ_s as n is lowered is well established theoretically [19] and experimentally for various 2DESs [17,18,20–23]. It occurs because of the increasing dominance of the Coulomb energy over the kinetic (Fermi) energy as n is decreased. To minimize its Coulomb energy, the 2DES tends to more easily align the electrons' spins so that they would spatially stay farther apart from each other. A similar mechanism is likely leading to the χ_ν enhancement. This is not unexpected, if we regard valley and spin as similar degrees of freedom. Indeed, in the standard effective mass approximation, where electrons near a semiconductor's conduction band minima are treated as quasiparticles with renormalized (i.e., *band* values) of parameters such as (the effective) mass and g factor but are otherwise free, spin, and valley are equivalent, SU(2) symmetric degrees of freedom. Interaction would then be expected to affect the spin and valley susceptibilities in a similar fashion. Interaction-
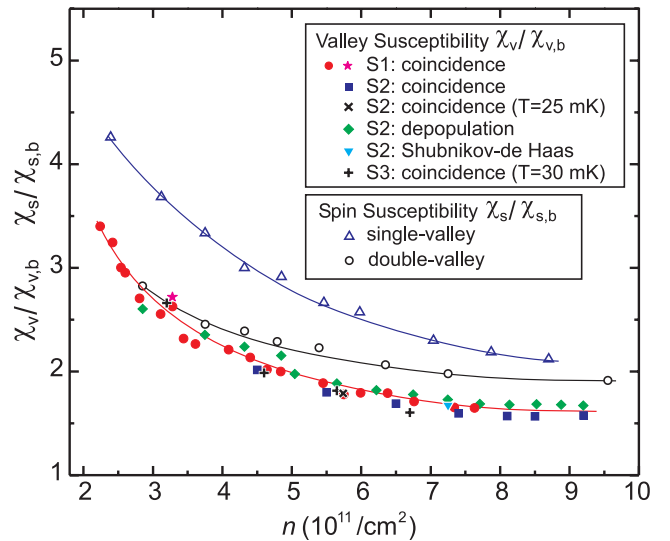

FIG. 2 (color online). Valley and spin susceptibilities, normalized to their band values, vs density. χ_ν was measured for three samples, S1, S2, and S3 using various techniques, as indicated. χ_s data were measured in several AlAs 2DES samples, including S2, and are shown for the case when the X and Y valleys are equally occupied (open circles), and for when only one valley is occupied (open triangles). Data points are from measurements at $T = 0.3$ K except as indicated. Lines are guides to the eye.

induced valley splitting, akin to exchange-induced spin-splitting, is in fact, known to occur in multivalley 2DESs in the presence of $B_\perp$ [24]. Evidence was also recently reported for interaction-induced quantum Hall valley Skyrmions, in direct analogy with spin Skyrmions [25].

Despite the similarities between the valley and spin degrees of freedom, there remains a puzzle. As shown in Fig. 2, within our experimental uncertainty, there is no spin polarization dependence of χ_v, although there is a valley polarization dependence of χ_s [18]. Our χ_v measurements are performed primarily at fixed $\nu \geq 4$ in a regime where the electrons are not fully spin polarized. But we have also measured χ_v at $\nu = 4$ in tilted magnetic fields, where the 2DES is fully spin polarized, and it agrees closely with χ_v in the unpolarized regime (see "star" data point in Fig. 2). However, as seen in Fig. 2, χ_s in a single valley system is larger than in the two-valley case [18]. This disparity may point to a fundamental difference between the valley and spin degrees of freedom, possibly reflecting the inadequacy of the effective mass approximation when one is dealing with interaction between electrons in different, *anisotropic* valleys located at different points of the Brillouin zone.

We close by making some general remarks. First, utilizing the spin degree of freedom to make functional devices (spintronics), or as a qubit for quantum computation, has been of much interest lately [1–3]. The valley degree of freedom may provide an alternative for such applications. For example, it may be possible to encode quantum information into the "valley state" of a two-level quantum dot. This type of qubit could potentially have much longer coherence times than those of the spin-based GaAs qubits where the electron spin coherence times suffer from a coupling of the electron and nuclear spins (see, e.g., Ref. [3]). The results reported here demonstrate that the valley populations can be modified *and* monitored, and provide values for one of the most basic parameters of the system, namely, its (valley) susceptibility. Second, the spin susceptibility of an interacting 2DES is expected to eventually diverge when n is lowered below a critical value at which the system attains a ferromagnetic ground state. There are also transitions to ferromagnetic quantum Hall states in samples with appropriate parameters. We believe that, under favorable conditions, e.g., at very small n or in the quantum Hall regime, "valley ferromagnetism" phenomena should also be prevalent. Finally, it is believed that there exists an intimate relationship between spin polarization and the 2D metal-insulator transition (MIT), and some have even suggested that the spin susceptibility diverges at the zero-field MIT critical density [21]. The similarity of spin and valley degrees of freedom suggests that the latter should play a role in the MIT problem as well. We have studied the MIT in the present system and our preliminary results indeed indicate that the 2DES can be driven to an insulating phase by increasing its valley polarization. A full account of our findings will be reported elsewhere.

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[4] These "band" values differ in general from the values for electrons in vacuum because of the electron's interaction with the host material's atoms.
[5] We define strain as $\epsilon = \epsilon_{[100]} - \epsilon_{[010]}$. With this definition, the splitting in X and Y valley energies is equal to $\epsilon E_{2,b}$.
[6] Our system's band parameters are: $m_b = \sqrt{m_l m_t} = 0.46 m_0$ where $m_l = (1.1 \pm 0.1)m_0$ and $m_t = (0.20 \pm 0.02)m_0$ are the longitudinal and transverse effective masses of AlAs [T. S. Lay *et al.*, Appl. Phys. Lett. **62**, 3120 (1993)]; $g_b = 2$, and $E_{2,b} = (5.8 \pm 0.1)$ eV [S. Charbonneau *et al.*, Phys. Rev. B **44**, 8312 (1991)].
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[10] Y. P. Shkolnikov *et al.*, Appl. Phys. Lett. **85**, 3766 (2004); note, however, that the applied strain was overestimated in that work because of a miscalibration.
[11] Determining χ_v from the onset of full valley polarization is analogous to measuring χ_s for 2DESs from the onset of full spin polarization using parallel magnetic field.
[12] The analog of this technique for the spin case is the determination of χ_s from measurements of spin subband densities as a function of parallel *and* perpendicular magnetic fields: $B_\perp$ induces Shubnikov–de Haas oscillations whose frequencies give the densities of the spin subbands while the total field causes the spin polarization; see, e.g., E. Tutuc *et al.*, Phys. Rev. Lett. **86**, 2858 (2001).
[13] In Fig. 2 we refer to these as "depopulation" and "Shubnikov–de Haas" data.
[14] This is equivalent to the coincidence technique of applying $B_\parallel$ and $B_\perp$ simultaneously to determine χ_s [F. F. Fang *et al.*, Phys. Rev. **174**, 823 (1968)].
[15] This fan diagram is qualitatively correct in the density range of interest where $0.9 \lesssim E_Z/E_C \lesssim 1.4$.
[16] Similarly, it has been reported in Ref. [17] that in samples with comparable densities to the ones studied here, the spin polarization is linear with magnetic field, i.e., χ_s does not depend on the degree of spin polarization.
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