## **Carrier-Density-Controlled Anisotropic Spin Susceptibility of Two-Dimensional Hole Systems**

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We study quantum-well-confined holes based on the Luttinger-model description for the valence band of typical semiconductor materials. Even when only the lowest quasi-two-dimensional (quasi-2D) subband is populated, the static spin susceptibility turns out to be very different from the universal isotropic Lindhard-function line shape obtained for 2D conduction-electron systems. The strongly anisotropic and peculiarly density-dependent spin-related response of 2D holes at long wavelengths should make it possible to switch between easy-axis and easy-plane magnetization in dilute magnetic quantum wells. An effective g factor for 2D hole systems is proposed.

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Introduction.-In semiconductors, electric current can be carried by conduction-band electrons or valence-band holes. Besides being oppositely charged, these two types of quasiparticles exhibit strikingly different magnetic properties. Band electrons are spin-1/2 particles like electrons in vacuum. In contrast, holes have a spin angular momentum of 3/2. Size quantization strongly affects the holes' spin-3/2 degree of freedom [1]. This is believed to be the origin of an unusual paramagnetic response observed for quantum-well-confined holes [2–4], and the same phenomenon is expected to stabilize out-of-plane easy-axis magnetism in a dilute-magneticsemiconductor (DMS) [5-7] two-dimensional (2D) hole system [8,9]. Controlling the confinement of holes thus enables appealing routes toward realizing magnetic semiconducting devices based on strain-induced anisotropies [10] or wave-function engineering in heterostructures [11,12]. Here we present a detailed theoretical study of the static spin susceptibility of 2D hole systems, which reveals unexpectedly rich magnetic properties of *p*-type quantum wells. The paramagnetic response is characterized by a strongly anisotropic and density-dependent effective g factor. In a 2D DMS system, valence-band mixing drives magnetic transitions that could enable new magnetoelectronic device functionalities [13,14] based on electric-field manipulation of the magnetization in low-dimensional systems.

*Background and aim.*—The magnetic properties of a many-particle system are most comprehensively characterized by the spin-susceptibility tensor [15]. In a homogeneous electron system with spin-rotational invariance, it has a universal isotropic form that depends only on the dimensionality of the system [16]. This case applies to the 2D electron systems realized by confining carriers from the conduction band in semiconductor heterostructures as long as inversion symmetry is not broken by the crystal lattice or due to structuring of the sample [17–20].

Here we consider the properties of 2D *hole* systems whose charge carriers have a spin-3/2 degree of freedom that is strongly coupled to their orbital motion even when inversion symmetry is intact [1]. We adopt the Luttinger model [21] in axial approximation [1,22,23], which provides a useful description of the uppermost valence band of typical semiconductors in situations where its couplings to the conduction band and split-off valence band are irrelevant. Subband *k*-dot-*p* theory [24,25] is employed to obtain the lowest quasi-2D hole subbands for a symmetric hard-wall confinement characterized by its spatial width *d*; see Fig. 1. We consider the case where only the lowest 2D subband is occupied and calculate the spin susceptibility. Based on this result, we discuss the paramagnetic response



FIG. 1. Lowest three (each of them doubly degenerate) subbands of a two-dimensional hole system realized by a symmetric hard-wall quantum-well confinement of width *d*. Dispersions are calculated based on the four-band Luttinger-model description of bulk valence-band states in axial approximation, using bandstructure parameters applicable to GaAs confined in [001] direction, and  $E_0 = -\pi^2 \hbar^2 \gamma_1 / (2m_0 d^2)$ . Gray lines indicate the range of energies and wave vectors for which only the lowest subband is occupied. This is the regime we focus on in this work.

of 2D holes and identify various magnetic phases that emerge in DMS quantum wells.

*Spin susceptibility of a quasi-2D system.*—The spin susceptibility of 2D charge carriers is given by [16]

$$\chi_{ij}(\mathbf{R}, z; \mathbf{R}', z') = -\frac{i}{\hbar} \int_0^\infty dt \, e^{-\eta t} \langle [S_i(\mathbf{R}, z; t), S_j(\mathbf{R}', z'; 0)] \rangle.$$
<sup>(1)</sup>

Here **R** and *z* are components of the position vector in the 2D (*xy*) plane and in the perpendicular (growth) direction, respectively. The spin density operator (in units of  $\hbar$ ) is defined in terms of field operators  $\Psi$ ,  $\Psi^{\dagger}$  and the Cartesian components  $\hat{J}_j$  of the charge carriers' intrinsic angular momentum as  $S_j(\mathbf{R}, z) = \Psi^{\dagger}(\mathbf{R}, z)\hat{J}_j\Psi(\mathbf{R}, z)$ . The field operators can be expressed in terms of operators associated with general eigenstates [labeled by band index *n* and inplane wave vector  $\mathbf{k} = (k_x, k_y)$ ] of the noninteracting Hamiltonian as  $\Psi(\mathbf{R}, z) = \sum_n \int \frac{d^2k}{(2\pi)^2} e^{i\mathbf{k}\cdot\mathbf{R}}\xi_{n\mathbf{k}}(z)c_{n\mathbf{k}}$ . The (normalized) spinors  $\xi_{n\mathbf{k}}(z)$  and eigenvalues  $E_{n\mathbf{k}}$  are obtained by solving the multiband Schrödinger equation for the confinement in growth direction of the 2D heterostructure. We can then express the spin susceptibility as

$$\chi_{ij}(\mathbf{R}, z; \mathbf{R}', z') = \int \frac{d^2 q}{(2\pi)^2} e^{i\mathbf{q}\cdot(\mathbf{R}-\mathbf{R}')} \chi_{ij}(\mathbf{q}; z, z') \quad (2a)$$

in terms of the 2D Fourier-transformed susceptibility

$$\chi_{ij}(\mathbf{q}; z, z') = \sum_{n,l} \int \frac{d^2k}{(2\pi)^2} \mathcal{W}_{ij}^{nl}(\mathbf{k}, \mathbf{q}; z, z')$$
$$\times \frac{n_F(E_{l\mathbf{k}+\mathbf{q}}) - n_F(E_{nk})}{E_{l\mathbf{k}+\mathbf{q}} - E_{nk} - i\hbar\eta}, \qquad (2b)$$

where  $n_F$  denotes the Fermi function, and

$$\mathcal{W}_{ij}^{nl}(\mathbf{k}, \mathbf{q}; z, z') = [\xi_{n\mathbf{k}}(z)]^{\dagger} [\hat{J}_i \xi_{l\mathbf{k}+\mathbf{q}}(z)] [\xi_{l\mathbf{k}+\mathbf{q}}(z')]^{\dagger} \times [\hat{J}_j \xi_{n\mathbf{k}}(z')].$$
(2c)

Axial symmetry of the 2D system implies  $E_{n\mathbf{k}} \equiv E_{nk}$ (where  $k \equiv |\mathbf{k}|$  and  $\phi_{\mathbf{k}}$  are the polar coordinates of  $\mathbf{k}$ ) and permits the ansatz [26,27]

$$\xi_{n\mathbf{k}}(z) = e^{-i\hat{J}_z\phi_{\mathbf{k}}}\bar{\xi}_{nk}(z),\tag{3}$$

simplifying calculation of the matrix elements  $\mathcal{W}_{ij}^{nl}$ . In the following, we consider the growth-direction-averaged spin susceptibility  $\bar{\chi}_{ij}(\mathbf{q}) = \int dz \int dz' \chi_{ij}(\mathbf{q}; z, z')$  calculated at zero temperature.

Luttinger-model description of quasi-2D holes.—Using the 4 × 4 Luttinger Hamiltonian in axial approximation for the bulk valence band, the bound states of 2D holes confined by a potential V(z) are given in terms of spinor wave functions  $\bar{\xi}_{nk}(z)$  that satisfy the Schrödinger equation  $[\mathcal{H}_0 + \mathcal{H}_1 + \mathcal{H}_2]\bar{\xi}_{nk} = E_{nk}\bar{\xi}_{nk}$ , where

$$\mathcal{H}_{0} = \frac{\hbar^{2}}{2m_{0}} \bigg[ \gamma_{1} \mathbb{1} - 2\tilde{\gamma}_{1} \bigg( \hat{J}_{z}^{2} - \frac{5}{4} \mathbb{1} \bigg) \bigg] \frac{d^{2}}{dz^{2}} + V(z), \qquad (4a)$$

$$\mathcal{H}_{1} = \frac{\hbar^{2}k}{m_{0}} \sqrt{2} \tilde{\gamma}_{2}(-i) (\{\hat{J}_{z}, \hat{J}_{-}\} + \{\hat{J}_{z}, \hat{J}_{+}\}) \frac{d}{dz}, \tag{4b}$$

$$\mathcal{H}_{2} = -\frac{\hbar^{2}k^{2}}{2m_{0}} \bigg[ \gamma_{1}\mathbb{1} + \tilde{\gamma}_{1} \bigg( \hat{J}_{z}^{2} - \frac{5}{4}\mathbb{1} \bigg) - \tilde{\gamma}_{3} (\hat{J}_{+}^{2} + \hat{J}_{-}^{2}) \bigg]. \quad (4c)$$

Here  $m_0$  is the electron mass in vacuum, hole energies are counted as negative from the bulk valence-band edge, and we used the abbreviations  $\hat{J}_{\pm} = (\hat{J}_x \pm i \hat{J}_y)/\sqrt{2}$ ,  $\{A, B\} = (AB + BA)/2$ . The constants  $\gamma_1$  and  $\tilde{\gamma}_j$  are materials-related band-structure parameters [28] and depend on the quantum-well growth direction.

Straightforward application of the subband k-dot-pmethod [24,25] yields the energy dispersions  $E_{nk}$  with associated eigenspinors  $\xi_{nk}$ . At k = 0, the eigenspinors are also eigenstates of  $\hat{J}_{\tau}$  with eigenvalues  $\pm 3/2$  (heavy holes) or  $\pm 1/2$  (light holes), which are split in energy. This phenomenon is often referred to as HH-LH splitting [1,22]. At finite k, the spinors  $\bar{\xi}_{nk}$  are not eigenstates of  $\hat{J}_z$  anymore. This phenomenon of HH-LH mixing arises because a spin-3/2 degree of freedom has a much richer structure than the more familiar spin-1/2 case [29]. Figure 1 shows the 2D-subband dispersions obtained for a symmetric hard-wall confinement of width d, using band-structure parameters for GaAs confined in the [001] direction. The numerically obtained  $E_{nk}$  and  $\xi_{nk}$  serve as input for the calculation of the 2D hole spin susceptibility according to Eq. (2) with Eq. (3).

Results for the 2D hole spin susceptibility.—Using polar coordinates  $(q, \phi_{\mathbf{q}})$  for the 2D wave vector  $\mathbf{q}$  and introducing the scale  $\chi_0 = 2m_0/(\gamma_1\hbar^2)$ , we find that the tensor elements of  $\bar{\chi}_{ij}(\mathbf{q})$  have the generic form

$$\bar{\chi}_{xx}(\mathbf{q}) = \chi_0 [F_{\parallel}(q) + G(q)\cos(2\phi_{\mathbf{q}})], \qquad (5a)$$

$$\bar{\chi}_{yy}(\mathbf{q}) = \chi_0 [F_{\parallel}(q) - G(q)\cos(2\phi_{\mathbf{q}})], \qquad (5b)$$

$$\bar{\chi}_{xy}(\mathbf{q}) = \chi_0 G(q) \sin(2\phi_{\mathbf{q}}), \tag{5c}$$

$$\bar{\chi}_{zz}(\mathbf{q}) = \chi_0 F_\perp(q). \tag{5d}$$

The functions  $F_{\parallel,\perp}(x)$  and G(x) depend on materials and morphological parameters of the 2D hole system, especially the hole sheet density  $n_{2D} \equiv k_F^2/(2\pi)$ , but G(0) = 0generally. Figure 2 shows typical results obtained at low, intermediate, and high densities where still only states in the lowest quasi-2D subband are occupied [30].

The density dependence of the 2D hole spin susceptibility follows a generic trend. In the limit of low density [Fig. 2(a)],  $\bar{\chi}_{zz}(\mathbf{q}) \gg \bar{\chi}_{xx}(\mathbf{q}) \sim \bar{\chi}_{yy}(\mathbf{q})$ , and the line shape of  $\bar{\chi}_{zz}(\mathbf{q})$  is similar to the universal 2D-electron (Lindhard) result [16]. The strong easy-axis response is expected [8,9,12] as a result of HH-LH splitting, which favors a spin-3/2 quantization axis perpendicular to the 2D plane [1]. Interestingly, the behavior of the spin susceptibility changes as density is increased. For intermediate values of



FIG. 2 (color online). **q**-dependent spin susceptibility for  $\phi_{\mathbf{q}} = 0$  and values of the 2D hole Fermi wave vector  $k_F$  as indicated in the individual panels. Calculations are based on the band structure shown in Fig. 1.

hole density, results like that shown in Fig. 2(b) are obtained, exhibiting *easy-plane* anisotropy in the long-wavelength limit and a nontrivial structure developing at wave vectors comparable to  $k_F$ . The significant deviation from both the universal-2D-electron behavior and also the easy-axis response expected from HH-LH *splitting* arises because, as the 2D hole density is increased, higher-k states get occupied that are more strongly influenced by HH-LH *mixing*. At the highest values of density where still only the lowest quasi-2D hole subband is populated, easy-axis anisotropy is restored in the long-wavelength limit but the

response at finite q becomes as important in strength as that for  $q \rightarrow 0$ .

Effective g factors for quasi-2D holes.—Motivated by recent experimental [2–4] and theoretical [31] interest in the paramagnetic response of 2D holes, we apply our results for the spin susceptibility of a noninteracting 2D hole system to define an effective g factor.

A magnetic field parallel to the *j* axis couples to the holes' spin via the Zeeman term  $\mathcal{H}_Z = 2\kappa\mu_B B_j \hat{J}_j$ , where  $\mu_B$  is the Bohr magneton and  $\kappa$  the bulk-hole *g* factor [1]. In the low-field limit, the paramagnetic susceptibility is given by  $\chi_{P,j} = (2\kappa\mu_B)^2 \bar{\chi}_{jj}(\mathbf{q} = 0)$  in terms of our calculated spin susceptibility [32]. Comparison of this relation with the expression for the Pauli susceptibility of conduction electrons from a parabolic band [16] suggests defining the effective *g* factor for a many-particle state via  $\chi_{P,j} \equiv (\frac{g_j \mu_B}{2})^2 \bar{\chi}_L(\mathbf{q} = 0)$ , which explicitly yields

$$g_j = 4\kappa \sqrt{\frac{\bar{\chi}_{jj}(\mathbf{q}=0)}{\bar{\chi}_L(\mathbf{q}=0)}}.$$
 (6)

Here  $\bar{\chi}_L$  is the static 2D hole Lindhard function; its  $\mathbf{q} = 0$  limit equals the density of states at the Fermi level. We find a density-dependent and anisotropic *g* factor, reflecting the interplay between HH-LH splitting and mixing in confined valence-band states. Figure 3 shows the density dependence of the transverse ( $\perp$ ) and in-plane ( $\parallel$ ) *g* factors for a 2D hole system. In the low-density limit, behavior expected from HH-LH splitting is found, whereas the paramagnetic response at intermediate and high density is substantially affected by HH-LH mixing.

2D-hole-mediated magnetism.—Magnetism is introduced into intrinsically nonmagnetic semiconducting materials via doping with magnetic ions [5–7] such as Mn, Co, Fe, or Gd. One way to generate an exchange interaction between any two localized magnetic moments embedded in a conductor is provided by the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism [15], which gives rise to the effective two-impurity spin Hamiltonian



FIG. 3 (color online). Effective g factor as a function of the 2D hole Fermi wave vector  $k_F$  for a perpendicular (dashed curve) and in-plane (solid curve) magnetic field.

$$\mathcal{H}_{\alpha\beta} = -G^2 \sum_{i,j} I_i^{(\alpha)} I_j^{(\beta)} \chi_{ij}(\mathbf{R}_{\alpha}, z_{\alpha}; \mathbf{R}_{\beta}, z_{\beta}).$$
(7)

Here  $I_i^{(\alpha)}$  denotes the *i*th Cartesian component of an impurity spin located at position ( $\mathbf{R}_{\alpha}, z_{\alpha}$ ) and *G* is the exchange constant for the contact interaction between the spin density of delocalized charge carriers with the impurity spins. For a random but on average homogeneous distribution of magnetic ions, standard mean-field theory [15] applied to the spin model described by the Hamiltonian of Eq. (7) yields the Curie temperatures

$$T_{C_j}^{(\mathrm{MF})} = 8\pi T_0 \frac{\bar{\chi}_{jj}(\mathbf{q}=0)}{\chi_0}$$
(8a)

for ferromagnetic order with magnetization direction parallel to the j axis. The temperature scale

$$T_0 = \frac{I(I+1)}{12} \frac{G^2}{k_B} \frac{n_I}{d} \frac{m_0}{\pi \hbar^2 \gamma_1}$$
(8b)

depends on the impurity-spin magnitude *I* and the average 3D density  $n_I$  of magnetic impurities, and its functional form is that obtained for 2D charge carriers in a parabolic band [9,33] with effective mass  $m_0/\gamma_1$ .

Figure 4 shows the mean-field Curie temperatures for perpendicular-to-plane ( $\perp$ ) and in-plane ( $\parallel$ ) magnetization directions calculated with the same input parameters used for obtaining the subbands given in Fig. 1. The density dependence of  $\bar{\chi}_{xx,zz}(0)$  is directly reflected in that of  $T_C^{\parallel,\perp}$ . At the mean-field level, the ordered state associated with the maximum transition temperature will be established. Our results suggest that the type of magnetic ordering can be modified by changing the 2D hole density, e.g., by adjusting the gate voltage in accumulationlayer devices [34]. Easy-axis magnetism prevails at low and high densities, whereas an unexpected easy-plane magnetic order emerges at intermediate values of the



FIG. 4 (color online). Mean-field Curie temperatures for 2D-hole-mediated easy-axis  $(T_C^{\perp})$  and easy-plane  $(T_C^{\parallel})$  magnetism as a function of the 2D hole Fermi wave vector  $k_F$ , obtained for a hard-wall confinement with width *d* and band-structure parameters applicable to GaAs.

density. Also for high densities, a local maximum appears in  $\bar{\chi}_{xx}(\mathbf{q})$  at  $q \approx 0.6k_F$ , which almost reaches the value of  $\bar{\chi}_{zz}(q=0)$ ; see Fig. 2(c). Even after averaging over the polar angle, we find that for  $k_F \sim 1.5\pi/d$  the in-plane susceptibility can have a maximum at  $q \neq 0$  which is as large as  $\bar{\chi}_{zz}(q=0)$ . Thus it may be possible that the highdensity easy-axis ferromagnetic state must coexist (or compete) with helical magnetism [15].

*Finite-temperature effects.*—Thermal excitation of spin waves (magnons) suppresses the magnitude of the magnetization below its mean-field value  $M_0$ . This effect is captured by the relation [35,36]

$$\frac{M(T)}{M_0} = 1 - \frac{1}{4\pi^2 n_I d} \int d^2 q \, n_{\mathbf{q}}(T), \tag{9}$$

where  $n_{\mathbf{q}}(T)$  is the occupation-number distribution function of magnon modes at temperature *T*. A spin-waverelated critical temperature is defined by the condition  $M(T_C^{(SW)}) = 0$  because, for  $T > T_C^{(SW)}$ , too many magnon excitations will have been excited to sustain a finite magnetization. In equilibrium,  $n_{\mathbf{q}}(T)$  is given by the Bose-Einstein distribution function  $n_B(\varepsilon_{\mathbf{q}}) = 1/(e^{\varepsilon_{\mathbf{q}}/[k_BT]} - 1)$ , which depends on the spin-wave energy dispersion  $\varepsilon_{\mathbf{q}}$ . The latter's expression in terms of the charge carriers'  $\mathbf{q}$ -dependent spin susceptibility depends on the type of magnetic order (Heisenberg, Ising, or helical) [35], but the parametrization

$$\varepsilon_{\mathbf{q}} = IG^2 \frac{n_I}{d} \chi_0 \bigg[ \bar{\varepsilon}_0 + \bar{c}_\nu \bigg( \frac{q}{k_F} \bigg)^\nu \bigg] \tag{10}$$

typically holds for the relevant energy range. The dimensionless quantities  $\bar{\varepsilon}_0$ ,  $\nu$ ,  $\bar{c}_{\nu}$  can be determined from the functional form of the q-dependent spin susceptibility. Stability of the magnetic order requires both coefficients  $\bar{\varepsilon}_0$  and  $\bar{c}_{\nu}$  to be positive. Specializing to our situation of 2D-hole-mediated magnetism, we see from Fig. 2(a) that the easy-axis magnetism expected at low hole-sheet densities is destabilized by magnons because  $\varepsilon_q < 0$ . Considering the easy-plane magnetism at intermediate densities, Fig. 2(b) reveals that the associated magnon dispersion is characterized by  $\nu = 2$  and  $\bar{\varepsilon}_0 = 0$ , which again implies destabilization of this magnetic order due to spin-wave excitations. For the easy-axis (Ising) magnet expected at high densities [cf. Fig. 2(c)], we find  $\nu = 2$ and  $\bar{\varepsilon}_0 > 0$ . In this case a finite spin-wave-related critical temperature is obtained. Further studies need to explore the effect of Coulomb interactions, which can stabilize ferromagnetic order mediated by 2D carriers [36].

*Conclusions.*—The spin susceptibility of 2D holes is strongly density dependent. In the low-density limit, the easy-axis response due to HH-LH splitting is exhibited. With increasing density, HH-LH mixing changes the spin-related response of confined holes even more drastically than the density response [37–39]. An effective g factor for 2D holes is proposed. We clarify the impact of

band-structure effects in 2D DMS systems that had previously been only considered for the 3D case [40-42] or outside the RKKY limit [43]. The switching behavior of the magnetization found here should be observable in *p*-type quantum wells where the 2D hole density is independently adjustable [34] and the magnetic doping is sufficiently low to ensure a sizable mean-free path.

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