ac-Driven Double Quantum Dots as Spin Pumps and Spin Filters

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We propose and analyze a new scheme of realizing *both* spin filtering and spin pumping by using ac-driven double quantum dots in the Coulomb blockade regime. By calculating the current through the system in the sequential tunneling regime, we demonstrate that the spin polarization of the current can be controlled by tuning the parameters (amplitude and frequency) of the ac field. We also discuss spin relaxation and decoherence effects in the pumped current.

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The emerging field of spintronics aims at creating devices based on the spin of electrons [1]. One of the most important requirements for any spin-based electronics is the ability to generate a spin current. Proposals for generating spin-polarized currents include spin injection by using ferromagnetic metals [2] or magnetic semiconductors [3]. Alternatively, one may use quantum dots (QDs) as spin filters or spin pumps [4,5]. For QD spin filters, dc transport through few electron states is used to obtain spin-polarized currents as demonstrated experimentally by Hanson *et al.* [6] following the proposal of Recher *et al.* [7]. Spin current rectification has also been realized [8]. The basic principle of spin pumps is, on the other hand, closely related to that of charge pumps. In a charge pump a dc current is generated by combining ac driving with either absence of inversion symmetry in the device or lack of time-reversal symmetry in the ac signal. The range of possible pumps includes turnstiles, adiabatic pumps, or nonadiabatic pumps based on photon-assisted tunneling (PAT) [9,10].

In this Letter we propose and analyze a new scheme of realizing *both* spin filtering and spin pumping by using a double quantum dot (DQD), in the Coulomb blockade regime, with time-dependent gate voltages and in the presence of a uniform magnetic field. The periodic variation of the gate potentials allows for a net dc current through the device even with no dc voltage applied [11,12]: if the system is driven at a frequency (or subharmonic) corresponding to the energy difference between two time-independent eigenstates, the electrons become completely delocalized [13,14]. If the left reservoir (chemical potential μ_L) can donate electrons to the left dot (at a rate Γ_L) and the right reservoir (chemical potential μ_R) can accept electrons from the right dot (at a rate Γ_R) the system will then pump electrons from left to right, even when there is no dc bias applied, namely $\mu_L = \mu_R$. Starting from this pumping principle our device has two basic characteristics: (i) if the process involves two-particle states, the pumped current can be completely spin-polarized *even if the contact leads are not spin-polarized* and (ii) the pumping can occur either through singlet [Fig. 1(a)] or triplet [Fig. 1(b)] states depending on the applied frequency, such that the *degree of spin polarization* can be tuned by means of the ac field. For example, if one drives the system (initially prepared in a state with $n = n_L + n_R = 3$ electrons: $|L = \downarrow\uparrow, R = \uparrow\rangle$) at a frequency corresponding to the energy difference between the singlets in both dots, the electron with spin $\downarrow$ becomes delocalized in the DQD system. If now the chemical potential for taking $\downarrow$ ($\uparrow$) electrons out of the right dot is above (below) μ_R, a spin-polarized current is

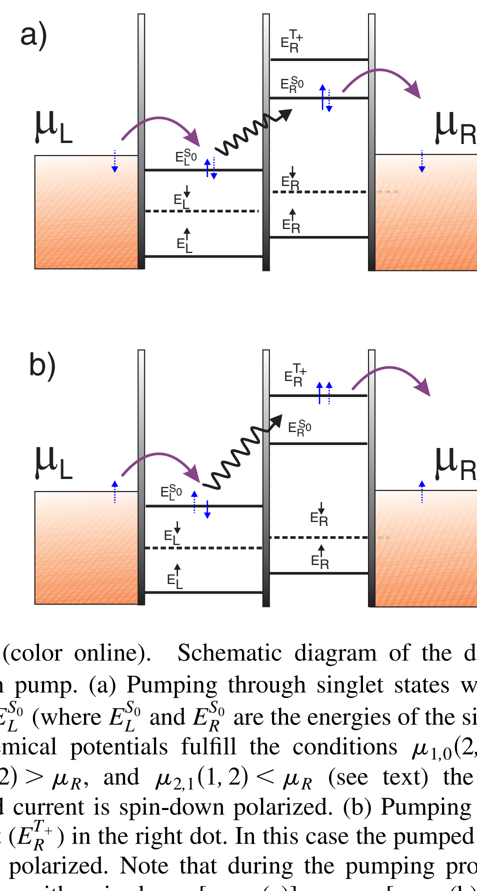

FIG. 1 (color online). Schematic diagram of the double dot electron pump. (a) Pumping through singlet states with $\hbar\omega = E_R^{S_0} - E_L^{S_0}$ (where $E_L^{S_0}$ and $E_R^{S_0}$ are the energies of the singlets). If the chemical potentials fulfill the conditions $\mu_{1,0}(2,1) < \mu_L$, $\mu_{2,0}(1,2) > \mu_R$, and $\mu_{2,1}(1,2) < \mu_R$ (see text) the resulting pumped current is spin-down polarized. (b) Pumping involving a triplet ($E_R^{T_+}$) in the right dot. In this case the pumped current is spin-up polarized. Note that during the pumping process only electrons with spin-down [case (a)] or -up [case (b)] become delocalized by the microwaves. Dashed arrows denote delocalized spins whereas solid arrows denote spins that remain localized on each dot.

generated. The above conditions for the chemical potentials can be achieved by breaking the spin-degeneracy through a Zeeman term $\Delta_z = g\mu_B B$, where B is the external magnetic field (which is applied parallel to the sample in order to minimize orbital effects), g is the effective g factor, and μ_B the Bohr magneton.

Our main findings can be summarized in Fig. 2 where we present a plot of the pumped current as a function of the applied frequency for a particular choice of parameters. Importantly, the current presents a series of peaks which are uniquely associated with a definite spin polarization: the pumped current is 100% spin-down (-up) polarized for $\omega = \omega_\downarrow$ ($\omega = \omega_\uparrow$). The other peaks correspond to subharmonics of the energy difference between the relevant states (see below). This particular example illustrates how by tuning the external ac field one can operate the driven DQD as a bipolar spin filter with no dc voltage applied.

Formalism.—Our system consists of an asymmetric DQD connected to two reservoirs kept at the chemical potentials μ_α, $\alpha = L, R$. Using a standard tunneling Hamiltonian approach, we write for the full Hamiltonian $\mathcal{H}_l + \mathcal{H}_{\mathrm{DQD}} + \mathcal{H}_T$, where $\mathcal{H}_l = \sum_\alpha \sum_{k_\alpha,\sigma} \epsilon_{k_\alpha} c^\dagger_{k_\alpha\sigma} c_{k_\alpha\sigma}$ describes the leads and $\mathcal{H}_{\mathrm{DQD}} = \mathcal{H}^L_{\mathrm{QD}} + \mathcal{H}^R_{\mathrm{QD}} + \mathcal{H}_{L\Leftrightarrow R}$ describes the DQD. It is assumed that only one orbital in the left dot participates in the spin-polarized pumping process whereas *two* orbitals in the right dot (energy separation $\Delta\epsilon$) have to be considered. The isolated left dot is thus modeled as a one-level Anderson impurity: $\mathcal{H}^L_{\mathrm{QD}} = \sum_\sigma E^\sigma_L d^\dagger_{L\sigma} d_{L\sigma} + U_L n_{L\uparrow} n_{L\downarrow}$, whereas the isolated

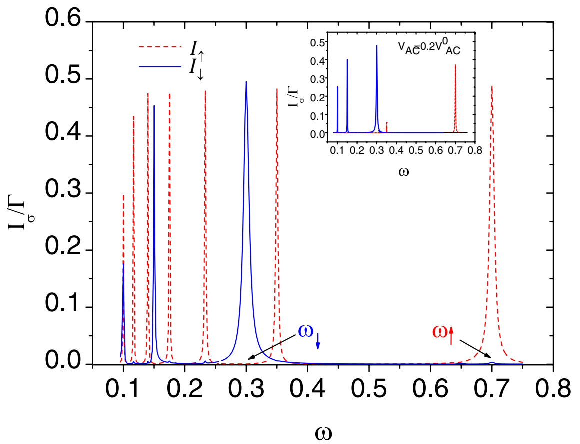

FIG. 2 (color online). Pumped current as a function of the ac frequency. The spin-down component (solid line) shows three peaks corresponding to one ($\omega = \omega_\downarrow = 0.3$), two ($\omega = 0.15$), and three ($\omega = 0.1$) photon processes, respectively. The spin-up component (dashed line) shows a main one-photon resonance at $\omega = \omega_\uparrow = 0.7$ and up to six more satellites corresponding to multiphoton processes. Parameters: $\Gamma_L = \Gamma_R = 0.001$, $t_{LR} = 0.005$, $U_L = 1.0$, $U_R = 1.3$, $J = 0.2$, $\mu_L = \mu_R = 1.31$, $\Delta_z = 0.026$, $\Delta\epsilon = 0.45$, $V_{\mathrm{ac}} = V^0_{\mathrm{ac}} = 0.7$ (in meV) correspond to typical values in GaAs QDs. In particular, the Zeeman splitting corresponds to a magnetic field $B \approx 1$ T. Inset: same as in main figure but with a lower intensity $V_{\mathrm{ac}} = V^0_{\mathrm{ac}}/5 = 0.14$.

right dot is modeled as $\mathcal{H}^R_{\mathrm{QD}} = \sum_{i\sigma} E^\sigma_{Ri} d^\dagger_{Ri\sigma} d_{Ri\sigma} + U_R(\sum_i n_{Ri\uparrow} n_{Ri\downarrow} + \sum_{\sigma,\sigma'} n_{R0\sigma} n_{R1\sigma'}) + J\mathbf{S}_0\mathbf{S}_1$. The index $i = 0, 1$ denotes the two levels. In practice, we take $E^\uparrow_L = E^\uparrow_{R0} = 0$ ($E^\downarrow_L = E^\downarrow_{R0} = \Delta_z$), so all the asymmetry is included in the charging energies $U_R > U_L$. Experimentally, this asymmetry can be realized by making the right dot smaller. $\mathbf{S}_i = (1/2)\sum_{\sigma\sigma'} d^\dagger_{Ri\sigma} \sigma_{\sigma\sigma'} d_{Ri\sigma'}$ are the spins of the two levels. As a consequence of Hund's rule, the intradot exchange, J, is ferromagnetic ($J < 0$) such that the singlet $|S_1\rangle = (1/\sqrt{2})(d^\dagger_{R0\uparrow} d^\dagger_{R1\downarrow} - d^\dagger_{R0\downarrow} d^\dagger_{R1\uparrow})|0\rangle$ is higher in energy than the triplets $|T_+\rangle = d^\dagger_{R0\uparrow} d^\dagger_{R1\uparrow}|0\rangle$, $|T_0\rangle = (1/\sqrt{2})(d^\dagger_{R0\uparrow} d^\dagger_{R1\downarrow} + d^\dagger_{R0\downarrow} d^\dagger_{R1\uparrow})|0\rangle$, and $|T_-\rangle = d^\dagger_{R0\downarrow} d^\dagger_{R1\downarrow}|0\rangle$. Because of the Zeeman splitting, $E^{T_-} > E^{T_0} > E^{T_+} = \Delta\epsilon + U_R - J/4$. Finally, we consider the case where $\Delta\epsilon > \Delta_z + J/4$ such that the triplet $|T_+\rangle$ is higher in energy than the singlet $|S_0\rangle = (1/\sqrt{2})(d^\dagger_{R0\uparrow} d^\dagger_{R0\downarrow} - d^\dagger_{R0\downarrow} d^\dagger_{R0\uparrow})|0\rangle$. $\mathcal{H}_{L\Leftrightarrow R} = \sum_{i,\sigma} t_{LR}(d^\dagger_{L\sigma} d_{Ri\sigma} + \mathrm{H.c.})$ describes tunneling between dots. The tunneling between leads and each QD is described by the perturbation $\mathcal{H}_T = \sum_{k_L,\sigma} V_L(c^\dagger_{k_L\sigma} d_{L\sigma} + \mathrm{H.c.}) + \sum_{i,k_R,\sigma} V_R(c^\dagger_{k_R\sigma} d_{Ri\sigma} + \mathrm{H.c.})$. $\Gamma_{L,R} = 2\pi\mathcal{D}_{L,R}|V_{L,R}|^2$ are the tunneling rates. It is assumed that the density of states in both leads $\mathcal{D}_{L,R}$ and the tunneling couplings are energy-independent.

We study the system by a reduced density matrix (RDM), $\rho = \mathrm{Tr}_L \chi$, where χ is the full density matrix and Tr_L is the trace over the leads. The dynamics of the RDM is formulated in terms of the eigenstates and eigenenergies of each isolated QD. We concentrate on the Coulomb blockade regime (with up to two electrons per dot, which defines a basis of 20 states) and study the sequential tunneling regime (Born-Markov approximation). For example, the diagonal elements of the RDM read

$$\dot{\rho}_{ss} = -\frac{i}{\hbar}[\mathcal{H}_{L\Leftrightarrow R}, \rho]_{ss} + \sum_{m\neq s} W_{sm}\rho_{mm} - \sum_{k\neq s} W_{ks}\rho_{ss}, \quad (1)$$

where W_{ij} are the transition rates. In addition, we consider an ac field acting on the dots, such that the single particle energy levels become $\epsilon_{L(R)} \rightarrow \epsilon_{L(R)}(t) = \epsilon_{L(R)} \pm \frac{eV_{\mathrm{ac}}}{2}\cos\omega t$, where eV_{ac} and ω are the amplitude and frequency, respectively, of the applied field. We include spin relaxation and decoherence phenomenologically in the corresponding elements of the equation for the RDM. Relaxation processes are described by the spin relaxation time $T_1 = (W_{\uparrow\downarrow} + W_{\downarrow\uparrow})^{-1}$, where $W_{\uparrow\downarrow}$ and $W_{\downarrow\uparrow}$ are spin-flip relaxation rates fulfilling a detailed balance. A lower bound for the spin relaxation time T_1 of 50 μs with a field $B = 7.5$ T has been obtained recently [15] for a single electron in a QD using energy spectroscopy and relaxation measurements. In the following, we focus on zero temperature results such that $W_{\downarrow\uparrow} = 0$ and thus $T_1 = W^{-1}_{\uparrow\downarrow}$. The rate T_2^{-1} (T_2 is the spin decoherence time) describes intrinsic spin decoherence. We take $T_2 = 0.1T_1$ in all the calculations [16].

In practice, we integrate numerically the dynamics of the RDM in the chosen basis. The dynamical behavior of the system is governed by rates which depend on the electrochemical potentials of the corresponding transitions. The electrochemical potential $\mu_{1(2),i}(N_1, N_2)$ of dot $L(R)$ is defined as the energy needed to add the $N_{1(2)}$th electron to energy level i of dot $L(R)$, while having $N_{2(1)}$ electrons on dot $R(L)$ [10]. The current from left to right is: $I_{L\to R}(t) = \sum_{s',s} W_{s's}\rho_{ss}(t)$, with a similar expression for $I_{R\to L}$. Here, states $|s\rangle$ are such that the right dot is occupied and $|s'\rangle$ states with one electron less. For ease in the notation, we take from now on $\hbar = e = 1$, such that $V_{\rm ac}$, ω, etc. have units of energy.

Results.—A calculation of the stationary current, for each direction of spin, namely $I_{\rm tot} = \sum_{\sigma=\uparrow,\downarrow} I_\sigma$ as a function of ω (and fixed intensity $V_{\rm ac} = V_{\rm ac}^0 = 0.7$), gives the results shown in Fig. 2. The main peak of $I_\downarrow$ (continuous line) occurs at $\omega = \omega_\downarrow \equiv E_{(\uparrow,\downarrow\uparrow)} - E_{(\downarrow\uparrow,\uparrow)} = U_R - U_L = 0.3$ (see caption) At this frequency $I_\uparrow \approx 0$ (dashed line), demonstrating the efficiency of the spin-polarized pump. For this particular case, pumping of spin-down electrons occurs as one electron with spin $\downarrow$ becomes delocalized (via a one-photon process) between both dots. If the chemical potential for taking $\downarrow$ electrons out of the right dot fulfills $\mu_{2,0}(1,2) = U_R - E_\uparrow > \mu_R$ while, on the other hand, the chemical potential for taking $\uparrow$ electrons out of the right dot fulfills $\mu_{2,1}(1,2) = U_R - E_\downarrow < \mu_R$, the resulting pumped current is spin-down polarized. We emphasize again that this pumping of spin-polarized ($\downarrow$) electrons is realized with unpolarized leads. Such spin-polarized current is obtained either through the sequence $(\downarrow\uparrow,\uparrow) \overset{\rm ac}{\Leftrightarrow} (\uparrow,\downarrow\uparrow) \overset{\Gamma_R}{\Rightarrow} (\uparrow,\uparrow) \overset{\Gamma_L}{\Rightarrow} (\downarrow\uparrow,\uparrow)$ or, alternatively, $(\downarrow\uparrow,\uparrow) \overset{\rm ac}{\Leftrightarrow} (\uparrow,\downarrow\uparrow) \overset{\Gamma_L}{\Rightarrow} (\downarrow\uparrow,\downarrow\uparrow) \overset{\Gamma_R}{\Rightarrow} (\downarrow\uparrow,\uparrow)$. The peaks at $\omega = \omega_\downarrow/N$ correspond to the absorption of N photons.

By increasing the frequency to $\omega = \omega_\uparrow \equiv E_{(\downarrow,\uparrow\uparrow)} - E_{(\downarrow\uparrow,\uparrow)} = \Delta\epsilon + U_R - U_L - J/4 = 0.7$ (see Fig. 2), a current peak with spin-up polarization appears. In this case, the pumped current occurs as one electron with spin $\uparrow$ becomes delocalized between the states $(\downarrow\uparrow,\uparrow)$ and $(\downarrow,\uparrow\uparrow)$. This spin-up electron subsequently decays to the right reservoir which leads to a pumped current through the sequence $(\downarrow\uparrow,\uparrow) \overset{\rm ac}{\Leftrightarrow} (\downarrow,\uparrow\uparrow) \overset{\Gamma_R}{\Rightarrow} (\downarrow,\uparrow) \overset{\Gamma_L}{\Rightarrow} (\downarrow\uparrow,\uparrow)$ or $(\downarrow\uparrow,\uparrow) \overset{\rm ac}{\Leftrightarrow} (\downarrow,\uparrow\uparrow) \overset{\Gamma_L}{\Rightarrow} (\downarrow\uparrow,\uparrow\uparrow) \overset{\Gamma_R}{\Rightarrow} (\downarrow\uparrow,\uparrow)$. At $\omega_\uparrow$, $I_\downarrow \approx 0$ such that the spin polarization, defined as

$$P(\omega, V_{\rm ac}) \equiv \frac{I_\uparrow - I_\downarrow}{I_\uparrow + I_\downarrow}, \tag{2}$$

has been completely reversed by tuning the frequency of the ac field, namely $P(\omega_\uparrow, V_{\rm ac}^0) = 1 = -P(\omega_\downarrow, V_{\rm ac}^0)$. Note that the energy difference between both processes, $\omega_\uparrow - \omega_\downarrow = \Delta\epsilon - J/4$ corresponds to the energy difference between the triplet excited state and the singlet ground state in the right dot *at zero magnetic field*.

Reducing the frequency, peaks corresponding to absorption of up to seven photons appear. Note that each of these peaks has a different width. This remarkable fact can be attributed to a renormalization of the interdot hopping induced by the ac potential [9,11]. At the frequency (or subharmonic) corresponding to the energy difference between two levels, the Rabi frequency becomes renormalized by the ac potential as $\Omega_{\rm Rabi} = 2t_{LR}J_N(V_{\rm ac}/\omega)$; J_N is the Bessel function of order N. The width of the peaks is given by the coupling to the leads provided that $\Gamma_{L,R} > \Omega_{\rm Rabi}$. By contrast, if $\Gamma_{L,R} < \Omega_{\rm Rabi}$ the width of the current peak is determined by $\Omega_{\rm Rabi}$. Since $\Omega_{\rm Rabi}$ depends on N in a nonlinear fashion (through the dependence of J_N on the ratio $V_{\rm ac}/\omega$), it follows that the widths of the peaks change in a nontrivial way as a function of ω. A similar nonlinear dependence of the height of the peaks as a function of the ratio $V_{\rm ac}/\omega$ is expected. We illustrate this nontrivial behavior in the inset of Fig. 2 where we explore the low intensities regime ($V_{\rm ac} = V_{\rm ac}^0/5 = 0.14$). In general, the trend we obtain is consistent with previous analytical estimations [11].

At frequencies where the one-photon process corresponding to pumping of $\downarrow$ electrons overlaps with multiphoton processes corresponding to pumping of $\uparrow$ electrons, the current is no longer fully spin-polarized. One can use this to modify the polarization of the current by changing $V_{\rm ac}$ (at fixed ω). We illustrate this with Fig. 3, where the parameters are chosen such that the $N = 1$ peak of $I_\downarrow$ is centered at the same frequency ($\omega = \omega_\downarrow = 0.3$) as the $N = 2$ peak of $I_\uparrow$. At this frequency, the spin polarization can be tuned by modifying $V_{\rm ac}$ (Fig. 3, inset). This result, together with those shown in Fig. 2, demonstrate that the spin polarization of the current $P(\omega, V_{\rm ac})$ can be fully manipulated by tuning either the frequency or the intensity of the external ac field.

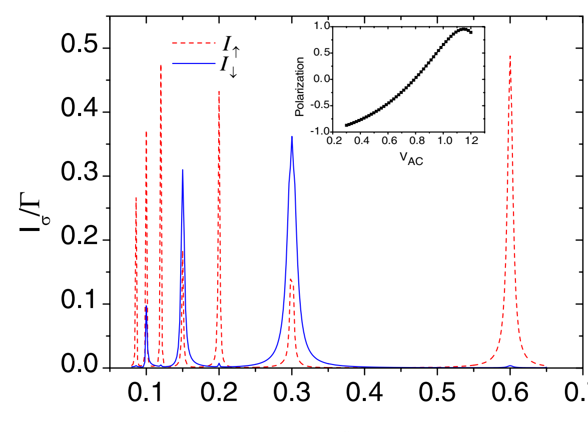

FIG. 3 (color online). Pumped current as a function of the ac frequency. Same parameters as Fig. 2 except $\Delta\epsilon = 0.35$. The interesting feature here is the overlap between the one-photon absorption peak of $I_\downarrow$ (solid line) and the two-photon absorption peak of $I_\uparrow$ (dashed line) at $\omega = \omega_\downarrow = 0.3$. The inset shows the spin polarization $P(\omega, V_{\rm ac})$ vs the ac intensity $V_{\rm ac}$ for fixed frequency $\omega = \omega_\downarrow = 0.3$ demonstrating the possibility of controlling the spin polarization of the current by tuning the intensity of the ac field.

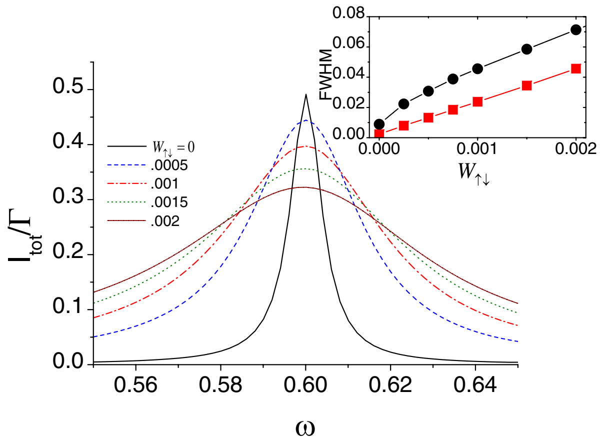

FIG. 4 (color online). Pumped current near resonance $\omega = \omega_\uparrow = 0.6$ for different relaxation rates. Inset: FWHM of the total current as a function of relaxation rate for strong (black dots) and weak (red squares) field intensity.

Finally, it is important to note also that, contrary to the case for spin-down pumping, the pumping of spin-up electrons leaves the double dot in the excited state $|\downarrow, \uparrow\rangle$. This makes the spin-up current extremely sensitive to spin relaxation processes. If the spin $\downarrow$ decays before the next electron enters into the left dot, namely, if $W_{\uparrow\downarrow} \gtrsim \Gamma_L$, a spin-down current appears through the cycle $(\downarrow\uparrow, \uparrow) \Leftrightarrow (\downarrow, \uparrow\uparrow) \overset{\Gamma_R}{\Rightarrow} (\downarrow, \uparrow) \overset{W_{\uparrow\downarrow}}{\Rightarrow} (\uparrow, \uparrow) \overset{\Gamma_L}{\Rightarrow} (\uparrow\downarrow, \uparrow)$ and the pumping cycle is no longer 100% spin-up polarized. We study this effect in Fig. 4, where we plot the main PAT peak at $\omega_\uparrow = 0.6$ for increasing $W_{\uparrow\downarrow}$. As one expects, the peak broadens as $W_{\uparrow\downarrow}$ increases. The full widths (FWHM) of the resonances are plotted as a function of $W_{\uparrow\downarrow}$ in the inset. For large intensities ($V_{ac} = V_{ac}^0$, circles) the FWHMs grow in a nonlinear fashion. This is reminiscent of the so-called saturation regime, a well known phenomenon in the context of optical Bloch equations. Note, however, that there are three energy scales involved now in the dynamics of the density matrix, Ω_{Rabi}, Γ, and $W_{\uparrow\downarrow}$, such that other sources of nonlinearity (like the ones described when discussing Fig. 2) cannot be completely ruled out [17]. In order to minimize nonlinear effects we investigate the low intensity regime ($V_{ac} = V_{ac}^0/5$, squares) where we expect a FWHM dominated by decoherence. The behavior is now linear with a slope which, interestingly, approaches FWHM $\sim 20 W_{\uparrow\downarrow} = 2/T_2$. Thus, experiments along these lines would complement the information about decoherence extracted from other setups [18].

Summary and experimental accessibility.—In summary, we have proposed and analyzed a new scheme of realizing *both* spin filtering and spin pumping by using ac-driven double quantum dots coupled to unpolarized leads. Our results demonstrate that the spin polarization of the current can be manipulated (including fully reversing) by just tuning the parameters of the ac field. Our results also show that the width in frequency of the spin-up pumped current gives information about spin decoherence in the quantum dot. We finish by mentioning that our proposal is within reach with today's technology for high-frequency experiments in quantum dots [10,13,14]. Indeed, PAT with two-electron spin states has recently been reported [19].

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