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Thermospin effects in a quantum dot connected to ferromagnetic leads

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We study a system composed of a quantum dot in contact with ferromagnetic leads held at different temperatures. Spin analogs to the thermopower and thermoelectric figure of merit are defined and studied as a function of junction parameters. It is shown that in contrast to bulk ferromagnets, the spin-thermopower coefficient in a junction can be as large as the Seebeck coefficient, resulting in a large spin figure of merit. In addition, it is demonstrated that the junction can be tuned to supply only spin current but no charge current. We also discuss experimental systems where our predictions can be verified.

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I. INTRODUCTION

Thermoelectricity—the relation between a temperature bias and a voltage bias—is a very old problem of solid-state physics. It has gained renewed interest in recent years[1–3] due to the prospect of utilizing nanostructures to develop high efficiency thermoelectric converters.[4,5] Theoretical models have been put forward for the thermoelectric transport through quantum point contacts,[6,7] quantum dots,[8–10] molecular junctions,[11] and other strongly correlated nanostructures.[12]

Recently, Uchida *et al.*[13] measured the spin equivalent of the charge-Seebeck coefficient, namely, the *spin*-Seebeck effect, in which a temperature difference between the edges of a bulk ferromagnetic (FM) slab induces a spin-voltage difference and generates spin current. The authors of Ref. 13 suggested using this effect to construct a spin-current source for spintronic devices.[14] However, the spin-Seebeck coefficient in this experiment was measured to be 4 orders of magnitude smaller than the charge-Seebeck coefficient. In addition, the temperature difference unavoidably generates also a regular voltage bias across the sample, which may preclude easy applicability in spintronic devices.

Here we study the *thermospin effect*, i.e., the spin analog to the Seebeck effect in a nanojunction composed of a quantum dot (either a molecule or a semiconductor quantum dot structure) placed between two FM leads. The charge transport properties of such systems have been studied both theoretically[15] and experimentally.[16,17] We define the spin analogs of the thermoelectric coefficients and show that in this particular case the spin- and charge-Seebeck coefficients are of the same order of magnitude. We also calculate the thermospin figure of merit (FOM) and show it to be relatively large, indicating high heat-to-spin-voltage conversion efficiency. Finally, it is demonstrated that the system parameters can be tuned such that a large spin current can be generated without any charge current, thus making this system ideal for spintronic applications.

II. DEFINITIONS OF SPIN-THERMAL COEFFICIENTS

Consider a system composed of some structure (for instance a quantum dot) placed between two FM leads, which we assume have the same magnetization alignment. The system is held at a temperature $T_L=T_R=T$. In linear response, the thermoelectric Seebeck coefficient is defined as minus the ratio between the voltage bias δV and the applied temperature bias δT that generates it (in the absence of charge current). In a spin system out of equilibrium one can define a spin-voltage bias as $\delta V_s=\Delta\mu_R-\Delta\mu_L$, where $\Delta\mu_\nu=\mu_{\nu\uparrow}-\mu_{\nu\downarrow}$, with $\mu_{s\nu}$ as the electrochemical potential of the spin s on either the right or left of the quantum dot. We expect this bias to be essentially zero when measured in the bulk of the FMs, but it may acquire a finite (albeit possibly small) value in proximity to the quantum dot.

To derive the spin-Seebeck coefficient, we consider a system in which there is both an infinitesimal temperature bias and spin-voltage bias. The charge and spin currents are defined as $I=I_\uparrow+I_\downarrow$, $I_s=I_\uparrow-I_\downarrow$, respectively (note that they have the same dimensions). In linear response, the spin current is given by $I_s=G_s\delta V_s+L_T\delta T$, where the response coefficient L_T is related to the fact that a temperature gradient can induce both a spin flow and an energy flow.[3,18] Setting $I_s=0$, we find the spin-Seebeck coefficient

$$S_s=-\frac{\delta V_s}{\delta T}=\frac{L_T}{G_s}. \tag{1}$$

Once S_s and G_s are defined, one may define a spin FOM, $Z_sT=\left|\frac{G_sS_s^2}{\kappa/T}\right|$, where $\kappa=\kappa_e+\kappa_{\rm ph}$ is the thermal conductance of the system, which has an electron contribution and a phonon contribution. The absolute value is taken because the spin conductance G_s may be negative. In analogy with charge transport, one expects that a system with $Z_sT>1$ is a good heat-to-spin-voltage converter.[1]

III. MODEL

The model consists of a quantum dot between two FM leads. The corresponding Hamiltonian of the system is

$$\mathcal{H}=\sum_{k,\nu=L,R}\sum_{s=\uparrow,\downarrow}(\varepsilon_{ks}-\mu_\nu)c_{\nu ks}^\dagger c_{\nu ks}+\sum_{s=\uparrow,\downarrow}\varepsilon_s d_s^\dagger d_s+U\hat{n}_\uparrow\hat{n}_\downarrow$$
$$+\sum_{k,\nu=L,R}\sum_{s=\uparrow,\downarrow}(\gamma_{\nu ks}c_{\nu ks}^\dagger d_s+\text{H.c.}), \tag{2}$$

where $c_{\nu ks}^\dagger$ creates an electron in the $\nu=L,R$ lead with spin s and energy ε_{ks} (the energy depends on spin due to the FM

splitting), $d_s^\dagger$ creates an electron in the dot with spin s, $\hat{n}_s = d_s^\dagger d_s$ is the number operator, U is the Coulomb charging energy, and ε_s is the energy level in the dot, which is spin dependent due to a field-induced Zeeman splitting, ΔB. The latter may originate from the magnetic field induced by the FM leads or by an external field. It may also arise from the presence of spin dipoles which are dynamically formed around a nanojunction.[3,19] $\gamma_{\nu ks}$ is the coupling between the leads and the dot. This is the simplest system that exemplifies the physics discussed in this paper but can also be realized in experiments.[16,17]

If the temperatures are higher than the Kondo temperature, in the sequential tunneling approximation (i.e., first order in $\gamma_{\nu ks}$) one can describe the system by using rate equations,[20] which describe the populations of the different states in the dot. The dot can be either empty (with probability P_0) or populated by a spin-up electron (P_1), by a spin-down electron (P_2), or by two electrons (P_3). The corresponding rate equation is

$$\frac{d}{dt}\begin{pmatrix} P_0 \\ P_1 \\ P_2 \\ P_3 \end{pmatrix} = \begin{pmatrix} -W_{0\to1}-W_{0\to2} & W_{1\to0} & W_{2\to0} & 0 \\ W_{0\to1} & -W_{1\to3}-W_{1\to0} & 0 & W_{3\to1} \\ W_{0\to2} & 0 & -W_{2\to3}-W_{2\to0} & W_{3\to2} \\ 0 & W_{1\to3} & W_{2\to3} & -W_{3\to1}-W_{3\to1} \end{pmatrix}\begin{pmatrix} P_0 \\ P_1 \\ P_2 \\ P_3 \end{pmatrix}. \tag{3}$$

The rates $W_{n\to n'}$ describe the probability per unit time to transfer from state n to state n'. They are evaluated by noting that the rate for an electron to hop onto (off) the dot is proportional to the probability $p_{\nu s}(\varepsilon)$ to find an electron (hole) in the ν lead with spin s at an energy ε. We assume that the coupling between the leads and the dot is energy independent (wide-band approximation) and for simplicity assume that the leads are symmetric [it is easy to show that our results, e.g., Eqs. (6) and (7), do not depend on junction asymmetry]. We thus have $p_{\nu s}(\varepsilon)=\alpha_s f_\nu(\varepsilon)$, where $f_\nu(\varepsilon)$ is the Fermi distribution of lead ν (with the corresponding temperature and chemical potential). The constant α_s parametrizes both the dot-lead coupling and the density of states (DOS), the spin dependence coming from both the FM band shift and the tunneling rate.[21] We assume that there is a majority of spin up in the ferromagnets (and that the leads magnetizations are aligned) and define $\gamma=\alpha_\downarrow/\alpha_\uparrow$. γ encodes the difference between both the DOS and the tunneling rates of the different spins.

Thus, for example, we have (setting $k_B=\hbar=1$, $\alpha_\uparrow=\alpha$, and $\alpha_\downarrow=\gamma\alpha$)

$$W_{0\to1} = \alpha[f((\varepsilon_\uparrow-\mu_L)/T_L)+f((\varepsilon_\uparrow-\mu_R)/T_R)],$$

$$W_{0\to2} = \alpha\gamma[f((\varepsilon_\downarrow-\mu_L)/T_L)+f((\varepsilon_\downarrow-\mu_R)/T_R)],$$

$$W_{1\to3} = \alpha\gamma[f((\varepsilon_\downarrow+U-\mu_L)/T_L)+f((\varepsilon_\downarrow+U-\mu_R)/T_R)],$$

$$W_{3\to2} = \alpha\{[1-f((\varepsilon_\uparrow+U-\mu_L)/T_L)] + [1-f((\varepsilon_\uparrow+U-\mu_R)/T_R)]\}, \tag{4}$$

and similarly for the rest of the transition rates. We assume that phonon-induced spin-relaxation processes in the dot are inhibited due to the presence of FM leads, and are hence slower than the spin transfer time scale, and may be neglected. We set the chemical potentials μ_L and μ_R as the zero of energy, and so the dot energies are $\varepsilon_{\uparrow,\downarrow}=\varepsilon\mp2\mu_B\Delta B$ (μ_B is the Bohr magneton). We will discuss two limiting cases of small and large Zeeman fields ΔB (in the sense that $2\mu_B\Delta B\gg\alpha$ or $2\mu_B\Delta B\ll\alpha$). The dot level ε may be tuned, e.g., by a gate voltage.

The steady-state solution is obtained by equating the right-hand side of Eq. (3) to zero. From this solution, one can determine the charge current, spin current, and heat current, using the continuity equation. For the charge and spin currents, one has $\frac{dn}{dt}=I_R-I_L$ and $\frac{dm}{dt}=I_{sR}-I_{sL}$, where n is the charge on the dot and $m=n_\uparrow-n_\downarrow$ is the magnetization of the dot. Using the rate equation one thus obtains (setting $e=\hbar=1$)

$$I_\nu = P_0(W^{(\nu)}_{0\to1}+W^{(\nu)}_{0\to2})-P_1(W^{(\nu)}_{1\to0}-W^{(\nu)}_{1\to3}) - P_2(W^{(\nu)}_{2\to0}-W^{(\nu)}_{2\to3})-P_3(W^{(\nu)}_{3\to1}+W^{(\nu)}_{3\to2}),$$

$$I_{s\nu} = P_0(W^{(\nu)}_{0\to1}-W^{(\nu)}_{0\to2})-P_1(W^{(\nu)}_{1\to0}+W^{(\nu)}_{1\to3}) - P_2(W^{(\nu)}_{2\to0}+W^{(\nu)}_{2\to3})-P_3(W^{(\nu)}_{3\to1}-W^{(\nu)}_{3\to2}), \tag{5}$$

where $W^{(\nu)}_{n\to n'}$ are scattering rates of transitions between the dot and the $\nu=L,R$ lead. Once all the currents are obtained, it is a matter of algebra to obtain the different transport coefficients using the linear response definition of the spin current.

IV. RESULTS

The procedure described above allows us to obtain analytic expressions for all the currents and thermoelectric/spin coefficients. The first result is that in the limit of $\Delta B\to0$, the charge-Seebeck coefficient S is independent of γ and is the same as was calculated in Ref. 10. The spin-Seebeck coefficient S_s is found to be proportional to S;

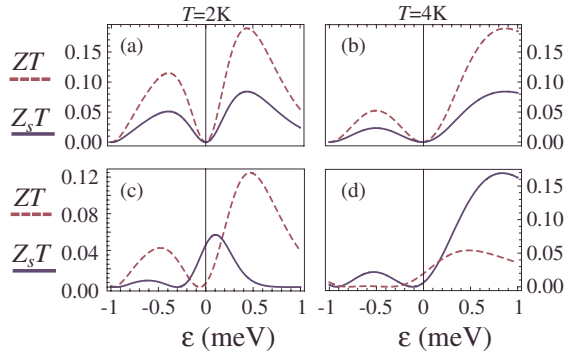

FIG. 1. (Color online) Spin FOM (Z_sT, solid line, purple online) and charge FOM (Z_T, dashed line, red online) as a function of ε at temperatures $T=2$ K (left column) and $T=4$ K (right column) for two values of Zeeman splitting: (a) and (b) $\Delta B=10^{-3}$ T and (c) and (d) $\Delta B=1.5$ T (see text for other numerical values). The spin and charge FOMs are comparable in size, and at certain parameters spin efficiency may even exceed that of charge.

$$S_s = \frac{1-\gamma}{1+\gamma} S. \tag{6}$$

Thus, for normal leads ($\gamma=1$) we have $S_s=0$, and for perfect FM leads ($\gamma=0$ or $\gamma=\infty$) the spin and charge coefficients are identical (up to a sign). Equation (6) shows that even for a moderate value of $\gamma=0.3$ we have $S_s \sim 0.5S$, as opposed to the bulk case where it is orders of magnitude smaller.[13] In the case of large ΔB, the situation is even more interesting since in fact S_s may become larger than S. In the limit of $U \to \infty$ and at $\varepsilon=0$ (i.e., the leads Fermi energies at the center of the Zeeman splitting) we find that

$$\frac{S_s}{S} = \frac{2}{1-\gamma \exp\left(\frac{2\mu_B \Delta B}{k_B T}\right)} - 1. \tag{7}$$

For a value of $\Delta B=1.5$ T at $T=5$ K, a value of $\gamma=0.3$ yields $\frac{S_s}{S} \approx 2.6$.

Let us now turn our attention to the FOM. We have calculated the FOM (spin and charge) numerically. For this we take the following parameters. The coupling between the dot and the leads is taken as $\alpha=10^{-2}$ meV (which is typical of semiconductor quantum dots). The charging energy is taken to be 2 orders of magnitude larger, $U=2$ meV, and we take $\gamma=0.2$. We add to the thermal conductance a phonon contribution which is $\kappa_{\rm ph}=3\kappa_0$ ($\kappa_0=\frac{\pi^2}{3}\frac{k_B^2}{h}T$ is the quantum of thermal conductance[22,23]), a reasonable value for nanoscale junctions.[24] In Fig. 1 we plot the spin FOM (Z_sT, solid line, purple online) and charge FOM (ZT, dashed line, red online) as a function of the dot energy level ε for two temperatures of 2 and 4 K (left and right columns, respectively) and for $\Delta B=10^{-3}$ T [Figs. 1(a) and 1(b)] and $\Delta B=1.5$ T [Figs. 1(c) and 1(d)]. The first value is a typical field produced by regular ferromagnets (e.g., iron), and the second corresponds to a large field splitting, which may be found in rare-earth ferromagnets or be induced by an external magnetic field.

From Fig. 1 one can see that the behavior of the spin and charge FOMs is similar for small Zeeman splitting and that

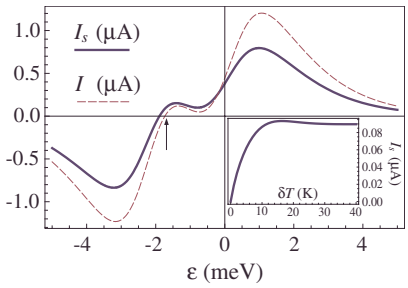

FIG. 2. (Color online) Spin current (solid line, purple online) and charge current (dashed line, red online) as a function of ε at a finite temperature difference $\delta T=10$ K (see text for other numerical parameters). The arrow indicates the energy at which the charge current vanishes and the spin current is finite. Inset: spin current as a function of temperature difference, evaluated at the energy ε at which the charge current vanishes.

Z_sT and ZT are of the same order of magnitude. The situation is different for large ΔB for which at certain energies close to $\varepsilon=0$ one may obtain small ZT but large Z_sT. This is due to the fact that the Zeeman splitting in that case preserves the particle-hole transport symmetry (the lack of which is responsible for charge thermopower) but dramatically changes the transport properties of different spins and, hence, increases the spin thermopower.

Finally, we study the system at finite currents. In the bulk, a temperature gradient will inevitably induce both charge and spin voltages,[13] and since the spin-Seebeck effect is much smaller than the charge-Seebeck effect, inducing large temperature biases (to generate sizeable spin currents) would result in even larger voltage biases. In the system studied here, one can instead tune the system parameters such that there will be a large spin current but a vanishing charge current.

In Fig. 2 the spin current I_s (solid line) and charge current I (dashed line) are plotted as a function of ε. Here the temperature $T=5$ K, $\Delta B=1.5$ T, and we have added a constant temperature gradient $\delta T=10$ K (the spin- and charge-voltage biases are zero). When the charge current vanishes (indicated by an arrow in Fig. 2) the spin current remains finite. The inset of Fig. 2 shows the dependence on the spin current, evaluated by varying the energy ε so that the charge current vanishes, as a function of the temperature bias δT. The magnitude of the spin current increases with the temperature difference and attains significant values for realizable temperature differences until it saturates at large temperatures (note, however, that the saturation temperature is comparable to the interaction energy U, and hence one expects that the sequential tunneling approximation breaks down at these temperatures). The finite spin current at large temperature difference stems from the fact that while the right lead is held at a high temperature, the temperature in the left lead is still low, allowing for differences in the tunneling rates of the different spins to be substantial. We stress that a situation of finite I_s but vanishing I cannot be achieved by using only a voltage bias, but a temperature bias is needed.

Our results are valid even if one considers additional single-particle levels in the dot. In the limit of infinite U, in fact, Eq. (6) is exact for the case of equidistant levels with no

Zeeman splitting. In the case with Zeeman splitting, we have numerically estimated S_s/S for up to five levels and found that even in the presence of the additional levels $S_s/S \sim 1$.

The ability to couple a quantum dot to FM leads[16,17] and to measure a local spin bias[13] has been demonstrated experimentally. It is thus reasonable that the results presented here are accessible by future experiments. Another interesting candidate for such experiments is graphene for which both the possibilities to fabricate quantum dots[25] and to bond FM leads to measure spin currents[26] have been demonstrated.

We also point out that if the leads are FM, extracting the spin current (or measuring the spin voltage) has to be done close to the junction at a distance shorter than the spin-diffusion length of the FM leads. Possible ways to circumvent this difficulty include the use of half-metallic leads (in which the spin-diffusion length should be very large) or to use a normal metal in contact with a thin FM layer for each lead, with the FM thin layers sandwiching the quantum dot.

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