

Thermoelectric Effect in Single-Molecule-Magnet Junctions

Rui-Qiang Wang,^{1,2} L. Sheng,² R. Shen,² Baigeng Wang,² and D. Y. Xing²

¹Laboratory of Quantum Information Technology, ICMP and SPTE, South China Normal University, Guangzhou 510006, China

²National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, Nanjing 210093, China

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We study the spin-dependent thermoelectric transport through a single-molecule-magnet junction in the sequential tunneling regime. It is found that the intrinsic magnetic anisotropy of the single-molecule magnet can lead to gate-voltage-dependent oscillations of charge thermopower and a large violation of the Wiedeman-Franz law. More interestingly, the spin-Seebeck coefficient is shown to be greater than the charge-Seebeck coefficient, and a pure spin thermopower or/and a pure spin current can be obtained by tuning only the gate voltage. It needs neither an external magnetic field or irradiation of circularly polarized light on the molecule nor ferromagnetic leads to realize these interesting effects, indicating the powerful prospect of single-molecule-magnet applications in spintronic devices.

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The research of high efficient heat-electricity conversion has become an active subject currently, and great interest is triggered in thermoelectric measurements of nanoscale junctions [1] due to developed nanotechnology. The thermoelectric transport coefficients can not only provide new insights into electronic transports, including Kondo correlations, vibrational excitations, and coherent effects [2], but also extract the information not accessible for electrical conductance measurements. For instance, it allows one to estimate the location of the Fermi energy [3] and even the chemical structure [4].

In nanostructures, the thermopower (or Seebeck coefficient) changes magnitude and sign alternately each time the Fermi level crosses the electron-hole symmetry point or resonant quantized levels, exhibiting oscillating behaviors with gate voltage [5,6]. The underlying physics is due to the Coulomb blockade effect. Coulomb interactions have also been shown to affect the efficiency of heat-electricity conversion remarkably, described in general by a dimensionless figure of merit. For macroscopic bulk samples, there is a Wiedeman-Franz (WF) law describing the universal relation between electrical conductance G and thermal conductance κ , i.e., Lorenz number $L = \kappa/GT = L_0$ with $L_0 = \frac{\pi^2}{3} (\frac{k_B}{e})^2$. Violation of the WF law is viewed as a mechanism to enhance the thermoelectric efficiency [7]. Recent reports showed that the Coulomb blockade effect can lead to strong violation, $L \gg L_0$, in a metallic single-electron transistor [8]. Large violation of the WF law has also been suggested in the molecular junctions and in the Luttinger liquids [9,10].

With the advancement of spintronics, the investigation on interplay of spin effects and heat transport has attracted much interest. The thermoelectric effects are shown to be dependent on the relative magnetic configurations of ferromagnetic leads [6,11]. Very recently, a new concept of the spin-Seebeck effect was proposed as an analog of the charge-Seebeck effect, and spin voltage generated by a temperature gradient was measured experimentally in a

ferromagnetic $\text{Ni}_{81}\text{Fe}_{19}$ film [12]. However, the measured spin thermopower in this bulk sample is so weak that it may be overwhelmed by the accompanied charge thermopower of several orders larger. Afterwards, Dubi and DiVentra [13] studied a quantum dot in contact with two ferromagnetic leads held at different temperatures and found that the spin thermopower can be as large as the charge thermopower and even can exceed the latter in magnitude. In their setup, the quantum dot is needed to be trapped between two ferromagnetic leads and subjected to a large Zeeman splitting, e.g., induced by a strong external magnetic field, which might limit the practical applications.

A single-molecule magnet (SMM), characterized by large local spin S (e.g., $S = 10$ for $\text{Mn}_{12}\text{-ac}$) and large intrinsic anisotropy, is a naturally magnetic system at low temperatures. Since 2006 when two groups [14] trapped an individual magnetic molecule in a nanogap to observe the transport spectra, considerable theoretical studies have focused on the spin-dependent electronic transport through the SMM, including the Kondo effect [15] and the magnetization reversion [16,17]. So far, however, there has been no report on thermal transport of the SMM junctions. It is expected that the study of the thermoelectric effect, especially the spin-relevant thermoelectric coefficients, in the SMM junctions will uncover some novel properties and provide a powerful tool of understanding the physics of magnetic excitations.

In this Letter, we study thermoelectric effects of the SMM junctions in the sequential tunneling regime. It is found that the intrinsic magnetic anisotropy, as a new type of mechanism, can give rise to oscillations of charge thermopower as well as a large violation of the WF law and that the spin-Seebeck coefficient of the SMM junction can be larger than its charge-Seebeck coefficient. More interestingly, even though the external magnetic field is absent and the electrodes are nonferromagnetic, one can generate thermally a pure spin thermopower and a pure spin current by tuning gate voltage.

Model.—Consider a magnetic molecular junction consisting of an SMM placed between two metallic electrodes in the presence of a temperature gradient δT . Its Hamiltonian includes the orbital and spin parts: $H = H_{\text{orbit}} + H_{\text{spin}}$, with

$$H_{\text{orbit}} = \sum_{\alpha k, \sigma} \varepsilon_{\alpha k \sigma} a_{\alpha k \sigma}^\dagger a_{\alpha k \sigma} + \sum_{\alpha k, \sigma} (t_\alpha a_{\alpha k \sigma}^\dagger c_\sigma + \text{H.c.}) + \sum_{\sigma} \varepsilon_0 \hat{n}_\sigma + U \hat{n}_\uparrow \hat{n}_\downarrow, \quad (1)$$

$$H_{\text{spin}} = -\frac{J}{2}(S_+ s_- + S_- s_+ + 2S_z s_z) - DS_z^2.$$

Here H_{orbit} is the Hamiltonian of a typical single-molecule transistor. $a_{\alpha k \sigma}^\dagger$ (c_σ^\dagger) is the creation operator for electrons in the $\alpha = L, R$ leads (on the molecular orbit) with spin σ and energy $\varepsilon_{\alpha k \sigma}$ (ε_0). $\hat{n} = c_\sigma^\dagger c_\sigma$ is the number operator, U is the Coulomb charging energy, and t_α is the energy-independent coupling between the leads and the SMM. H_{spin} is the spin Hamiltonian of the SMM with a giant local spin \mathbf{S} and a magnetic uniaxial anisotropy constant D [16]. J represents the Hund's exchange coupling between the SMM spin \mathbf{S} and electron spin $\mathbf{s} = \sum_{\sigma \sigma'} c_{\sigma'}^\dagger (\tau_{\sigma \sigma'} / 2) c_{\sigma'}$ with $\tau_{\sigma \sigma'}$ the Pauli matrix elements. $S_\pm = S_x \pm iS_y$ (similarly for s_\pm) are spin-raising operators and S_z (s_z) is the z component of the spin operator, where the z direction is chosen along the magnetic easy axis. The magnetic anisotropy perpendicular to the easy axis is shown to be smaller than D by several orders in magnitude and so is disregarded generally in H_{spin} [16].

It is assumed that the transport is dominated by the sequential tunneling through the SMM level, and the co-tunneling and direct tunneling are so weak that they can be neglected safely. We start with the Liouville–von Neumann equation and obtain a set of master equations in terms of the reduced density matrix. For the weak coupling of the molecule dot, the master equation reduces to be a set of rate equations [16,17]:

$$\frac{\partial P^i}{\partial t} = \sum_f (P^f W_{f \rightarrow i} - P^i W_{i \rightarrow f}). \quad (2)$$

Here P^i is the probability of finding electrons in the many-body state $|i\rangle$ of the SMM, and $W_{i \rightarrow f}$ is the transition rate between initial state $|i\rangle$ and final state $|f\rangle$. The transition rates can be written as a sum over the two leads and the two spin channels: $W_{i \rightarrow f} = \sum_{\alpha, \sigma} (W_{i \rightarrow f}^{\alpha \sigma +} + W_{i \rightarrow f}^{\alpha \sigma -})$, where $W_{i \rightarrow f}^{\alpha \sigma +} = \Gamma_{\alpha \sigma} f_\alpha^+ (\epsilon_f - \epsilon_i - \mu_\alpha) |\langle f | c_\sigma^\dagger | i \rangle|^2$ stands for the transition rate from lead α to the SMM and $W_{i \rightarrow f}^{\alpha \sigma -} = \Gamma_{\alpha \sigma} f_\alpha^- (\epsilon_f - \epsilon_i - \mu_\alpha) |\langle i | c_\sigma^\dagger | f \rangle|^2$ indicates that from the SMM to lead α . $f_\alpha^+ = 1/[1 + e^{(\varepsilon_\sigma - \mu_\alpha)/k_B T_\alpha}]$ is the Fermi-Dirac distribution, and $f_\alpha^- = 1 - f_\alpha^+$, with T_α and μ_α as the temperature and chemical potential in lead α , respectively. $\Gamma_{\alpha \sigma} = 2\pi \rho_{\alpha \sigma} |t_\alpha|^2$ characterizes the level-width function with $\rho_{\alpha \sigma}$ as the density of states at the Fermi level of lead α .

The next step is to diagonalize Hamiltonian H_{spin} , as done in Ref. [16]. The eigenstates fall into sectors with $n = 0, 1, 2$ electrons in the SMM, and another good quantum number is the magnetic quantum number m , which is the eigenvalue of the z component of the total spin, $S_z + s_z$. As a result, the SMM eigenstates and eigenvalues are labeled with $|i\rangle = |n, m\rangle$ and $\epsilon(n, m)$, respectively. From factor $|\langle f | c_\sigma^\dagger | i \rangle|^2$ in $W_{i \rightarrow f}$, it follows that there are basic selection rules: $|\Delta n| = 1$ and $|\Delta m| = 1/2$. Specifically, we obtain the transition rates for spin up as

$$W_{(0,m) \Rightarrow [1, m+(1/2)]}^{\alpha \uparrow \pm} = \Gamma_{\alpha \uparrow} f_\alpha^\pm \left[\epsilon\left(1, m + \frac{1}{2}\right) - \epsilon(0, m) - \mu_\alpha \right] \times |B_{m+(1/2)}|^2,$$

$$W_{[1, m-(1/2)] \Rightarrow (2, m)}^{\alpha \uparrow \pm} = \Gamma_{\alpha \uparrow} f_\alpha^\pm \left[\epsilon(2, m) - \epsilon\left(1, m - \frac{1}{2}\right) - \mu_\alpha \right] \times |A_{m-(1/2)}|^2. \quad (3)$$

Similar expressions for spin down can be obtained from Eq. (3) by making an exchange between $+\frac{1}{2}$ and $-\frac{1}{2}$ and that between A_m and B_m . Here the eigenenergies and the Clebsch-Gordan coefficients A_m and B_m for the coupling of spin angular moments \mathbf{s} and \mathbf{S} can be found in Ref. [16]. As SJ is chosen large enough, the higher branch $\epsilon^+(1, m)$ is about SJ higher than the lower branch $\epsilon^-(1, m)$ and can be neglected [17].

By setting the left-hand side of Eq. (2) to zero and using the continuity condition, one can obtain the formula for spin-resolved current $I_\alpha^\sigma = \frac{-e}{h} \sum_{i, f} (W_{i \rightarrow f}^{\alpha \sigma +} - W_{f \rightarrow i}^{\alpha \sigma -}) P^i$, so that the charge current can be evaluated by $I_c^\alpha = I_\alpha^\uparrow + I_\alpha^\downarrow$ and the spin current by $I_s^\alpha = I_\alpha^\uparrow - I_\alpha^\downarrow$. Similarly, the electronic heat flux from lead α into the molecule is derived with the fundamental thermodynamics, $I_Q^\alpha = \frac{d}{dt} \langle H_\alpha \rangle - \mu_\alpha \frac{d}{dt} \langle \sum_{k, \sigma} a_{\alpha k \sigma}^\dagger a_{\alpha k \sigma} \rangle$, yielding

$$I_Q^\alpha = \frac{1}{h} \sum_{\sigma, i, f} (\epsilon_f - \epsilon_i - \mu_\alpha) [W_{i \rightarrow f}^{\alpha \sigma +} - W_{f \rightarrow i}^{\alpha \sigma -}] P^i. \quad (4)$$

The thermoelectric coefficients are defined from the interplay of thermal and electrical properties. In linear response with respect to bias-voltage difference $\delta\mu = \mu_L - \mu_R$ and temperature gradient $\delta T = T_L - T_R$ across the system, the charge and heat currents are given by ($e = h = 1$) $I_c = \mathcal{L}_0 \delta\mu + \frac{\mathcal{L}_1}{T} \delta T$ and $I_Q = -\mathcal{L}_1 \delta\mu - \frac{\mathcal{L}_2}{T} \delta T$, respectively. Thus, one can determine a set of charge thermoelectric coefficients by taking $I_c = 0$: thermal conductance $\kappa = \frac{1}{T} (\mathcal{L}_2 - \mathcal{L}_1^2 / \mathcal{L}_0)$ and charge thermopower $S_c = \frac{-\mathcal{L}_1}{T \mathcal{G}_c}$ with charge conductance $G_c = \mathcal{L}_{0\uparrow} + \mathcal{L}_{0\downarrow}$. Similarly, a temperature gradient can induce both a spin flow and an energy flow. Under the condition of $I_s = 0$, the vanishing spin current induced by spin bias δV_{spin} and δT yields the spin thermopower [13] $S_s = \frac{-\mathcal{L}_1}{T \mathcal{G}_s}$, with $G_s = \mathcal{L}_{0\uparrow} - \mathcal{L}_{0\downarrow}$.

Results.—In this section, we perform numerical calculations for Mn₁₂-ac with typical parameters [14]: $S = 10$

and $D = 0.06$ meV with symmetric coupling, $\Gamma_{\alpha\sigma} = \Gamma_0$, independent of spin. At low temperatures, chosen to be $T = 1$ K, the electrons in spin states cannot energetically overcome the high anisotropy-induced energy barrier (DS^2) to enter into the opposite spin states, so that they always stays at the potential well with $m \geq 0$ prepared originally.

In Fig. 1(a), the charge-Seebeck coefficients S_c are plotted as a function of dot energy level ε_0 (or gate voltage). For $D = J = 0$, S_c exhibits linear behavior, which is just the result for the usual molecular junction with vanishing U in the sequential tunneling [9]. Interestingly, for finite D and J , one finds an oscillation of S_c around $S_c = 0$ at $\varepsilon_0 = 0$, two maxima of the absolute value of S_c with ε_0 deviated positively and negatively from the zero point, and two S_c vs ε_0 straight lines across the abscissa of $S_c = 0$. The change in sign is associated with the competition of the heat fluxes carried by electron-type flow ($\varepsilon_0 < 0$) and by hole-type flow ($\varepsilon_0 > 0$). The three points of $S_c = 0$ correspond to two resonant peaks of electrical conductance and the midpoint between the peaks.

To clarify this point, we further derive the analytic formula for the thermopower by taking into account only transitions between the three lowest energy states $(1, S + \frac{1}{2})$, $(0, S)$, and $(2, S)$, yielding

$$S_c = -\frac{k_B}{e} \frac{\varepsilon_{1,0} e^{\varepsilon_{2,0}} + \varepsilon_{2,0} e^{\varepsilon_{1,0}} + \varepsilon_{2,1}}{1 + 2e^{\varepsilon_{1,0}} + e^{\varepsilon_{2,0}}}, \quad (5)$$

with $\varepsilon_{1,0} = \frac{1}{k_B T} [\varepsilon(1, S + \frac{1}{2}) - \varepsilon(0, S)]$, $\varepsilon_{2,1} = \frac{1}{k_B T} \times [\varepsilon(2, S) - \varepsilon(1, S + \frac{1}{2})]$, and $\varepsilon_{2,0} = \frac{1}{k_B T} [\varepsilon(2, S) - \varepsilon(0, S)]$. The corresponding result is shown by the dashed line in Fig. 1(a), which is well consistent with the numerical result

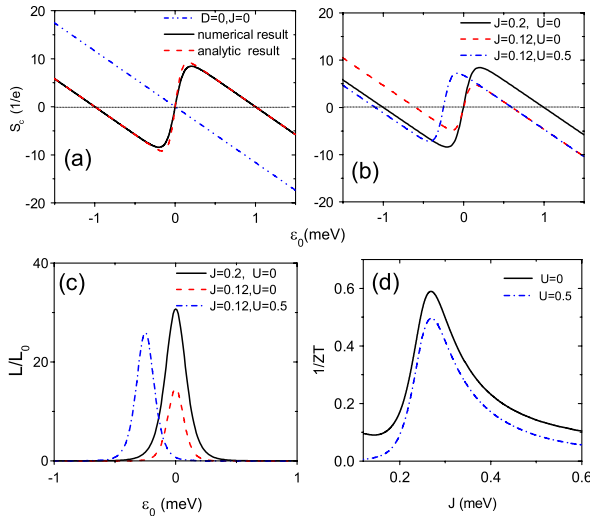


FIG. 1 (color online). (a) Charge thermopower S_c for non-magnetic molecule ($D = J = 0$) and SMM ($D = 0.06$ meV, $J = 0.2$ meV) with $U = 0$; dashed lines: analytic result from Eq. (5); solid lines: numerical result. (b) S_c and (c) Lorenz number L/L_0 for different J and U ; as functions of molecular level ε_0 . (d) Variation of the figure of merit ZT with J for $\varepsilon_0 = 0.2$.

above (solid line). This indicates that at low temperature, the main contribution to the thermopower originates from these lowest energy states. At $\varepsilon_0 = 0$, we have $\varepsilon(0, S) = \varepsilon(2, S)$, and from Eq. (5), it follows that $S_c = 0$. In this case, the heat flux due to the transition between states $(1, S + \frac{1}{2})$ and $(0, S)$ is compensated by that due to the transition between $(1, S + \frac{1}{2})$ and $(2, S)$, they having the equal rate but opposite directions. If denoting $\delta = \varepsilon_{1,0}$ and $u_{\text{eff}} = \varepsilon_{2,1} - \varepsilon_{1,0}$, one finds that Eq. (5) resembles in form the formula for thermopower with finite U , e.g., Eq. (19) in Ref. [9]. Here u_{eff} behaves as an effective Coulomb interaction, and $\varepsilon_0 = -\frac{u_{\text{eff}}}{2}$ ($\delta = -u_{\text{eff}}$) and $\varepsilon_0 = \frac{u_{\text{eff}}}{2}$ ($\delta = 0$) act as two resonant levels in the Coulomb blockade effect [6,9]. However, it is emphasized that u_{eff} is of different physics origin, induced purely by magnetic parameters (J , S , and D). Moreover, u_{eff} and δ are dependent on each other, $\delta = \varepsilon_0 - \frac{u_{\text{eff}}}{2}$, which makes the midpoint of $S_c = 0$ fixed at $\varepsilon_0 = 0$, regardless of magnetic parameters. In the presence of U , the thermopower zero points located originally at $\varepsilon_0 = 0$ and $-\frac{u_{\text{eff}}}{2}$ are shifted to $\varepsilon_0 = -\frac{U}{2}$ and $-\frac{u_{\text{eff}}}{2} - U$, respectively, but the zero point at $\varepsilon_0 = \frac{u_{\text{eff}}}{2}$ remains unchanged, as shown in Fig. 1(b). The combination of the magnetic parameters and Coulomb effect affects the magnitude of S_c via an effective Coulomb repulse parameter $u_{\text{eff}} + U$. Calculations also show that, with increasing temperature, the maxima of the absolute value of S_c decays significantly and the thermopower oscillations are suppressed due to exponential dependence of temperature, as in Eq. (5).

In Fig. 1(c), we plotted the Lorenz number L/L_0 as a function of energy level ε_0 , where $L \gg L_0$ around the symmetry point implies a large violation of the WF law. Even at $U = 0$, the large violation still appears, purely as a consequence of the spin-selection transitions between intrinsic magnetic states. The inverse figure of merit, defined as $ZT = G_c S_c^2 T / \kappa$, is plotted as a function of spin exchange energy J in Fig. 1(d). A large value of ZT indicates a high heat-electricity efficiency for the SMM devices. Figures 1(b)–1(d) also show the variation of S_c , L/L_0 , and ZT with J and U , in which the enhancement of them can be achieved by adjusting appropriate parameters.

In what follows, we focus on the spin-Seebeck effect in the magnetic molecule system. The charge thermopower S_c and the spin thermopower S_s are plotted in Fig. 2(a) and their ratio S_c/S_s in Fig. 2(b). Interestingly, it is shown that, in the vicinity of $\varepsilon_0 = 0$, S_s can exceed S_c greatly, i.e., $|S_s/S_c| \gg 1$. Especially at $\varepsilon_0 = 0$, where $S_c = 0$, there is only a spin-Seebeck coefficient but no charge counterpart, indicating that a pure spin thermopower can be induced by temperature gradient across the SMM device. With ε_0 departing away from $\varepsilon_0 = 0$, S_s becomes very close to S_c , keeping $|S_c/S_s| \approx 1$. The whole curve in Fig. 2(b) can be fitted approximatively to the formula $S_c/S_s = (e^{\varepsilon_{2,0}} - 1)/(1 + 2e^{\varepsilon_{1,0}} + e^{\varepsilon_{2,0}})$.

In order to understand the underlying physics, we plot the spin-dependent currents in Fig. 2(c) as functions of ε_0 .

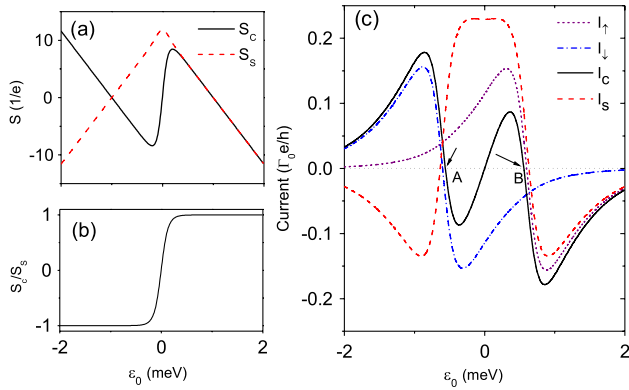


FIG. 2 (color online). (a) Charge thermopower S_c and spin thermopower S_s , (b) their ratio S_c/S_s , and (c) spin-dependent currents $I_{\uparrow(\downarrow)}$, charge current I_c , and spin current I_s , as functions of molecular level ϵ_0 .

At $\epsilon_0 = 0$, the largest occupation probability is at state $(1, S + \frac{1}{2})$ because $\epsilon(0, S) = \epsilon(2, S) > \epsilon(1, S + 1/2)$. According to the spin-selection rule, the up-spin electron transition is dominated by $(1, S + \frac{1}{2}) \rightarrow \epsilon(0, S)$ and down-spin electron transition by $(1, S + \frac{1}{2}) \rightarrow \epsilon(2, S)$. The former is responsible for electron ejection out of the dot (from right to left), while the latter is responsible for electron injection onto the dot (from left to right). The resulting current I_\uparrow and I_\downarrow have the same magnitude but opposite directions, which makes the charge current $I_c = I_\uparrow + I_\downarrow$ vanishing but the spin current reach its maximum, resulting in a pure spin current $I_s = I_\uparrow - I_\downarrow$, as shown in Fig. 2(c). In this case, the heat flux is generated mainly by the transferred charge, and so the heat conductance roughly follows the electrical conductance. Since up-spin and down-spin electrons carry exactly the same amount of heat but move along the opposite directions, the pure spin-Seebeck coefficient can be obtained at $\epsilon_0 = 0$, as shown in Figs. 2(a) and 2(b). For $\epsilon_0 < 0$, we have $\epsilon(0, s) > \epsilon(1, s + 1/2) > \epsilon(2, s)$, so the down-spin transition $|2, s\rangle \rightarrow |1, s + 1/2\rangle$ dominates the transport and the charge current is close to I_\downarrow roughly. The situation is just the opposite for $\epsilon_0 > 0$, in which the charge current is close to I_\uparrow roughly. It then follows that $S_c/S_s = 1$ for $\epsilon_0 > 0$ and $S_c/S_s = -1$ for $\epsilon_0 < 0$. In addition, it is found that the pure spin current can also be generated at indicated points A and B in Fig. 2(c), but no pure spin thermopower appears there, as shown in Figs. 2(a) and 2(b). Here the heat conductance no longer simply follows the electrical conductance for non-zero ϵ_0 , because the heat transfer is essentially the energy transport which is greatly relevant to the energies of the incident electrons.

Summary.—In summary, the SMM model is applied to study the linear-response thermoelectric effect of magnetic molecules in the sequential tunneling regime. At low temperatures, the intrinsic magnetic anisotropy of SMMs can lead to oscillations of charge thermopower with gate voltage and a large violation of the WF law, behaving similarly

to the Coulomb blockade effect. Most interestingly, it is found that the spin-Seebeck coefficient in SMMs can be greater than the charge-Seebeck coefficient, and even a pure spin thermopower can be obtained accompanied by the charge thermopower vanishing. This spin-Seebeck effect is attributed to the spin-dependent transitions governed by the spin-selection rule associated with the intrinsic magnetic states. These spin-related behaviors cannot be induced by the Coulomb blockade effect. In the present SMM device, it needs neither an external magnetic field nor ferromagnetic leads to generalize thermally the pure spin current or/and pure spin thermopower, which is quite different from in the nonmagnetic molecular device [13]. It is expected that the present study of the SMM thermoelectricity is helpful in the design of novel spintronic devices, e.g., a heat-spin converter.

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