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Unusual low-temperature thermopower in the one-dimensional Hubbard model

C. A. Stafford

Center for Superconductivity Research, Department of Physics, University of Maryland, College Park, Maryland 20742

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The low-temperature thermoelectric power of the repulsive-interaction one-dimensional Hubbard model is calculated using an asymptotic Bethe ansatz for holons and spinons. The competition between the entropy carried by the holons and that carried by the backflow of the spinons gives rise to an unusual temperature and doping dependence of the thermopower which is qualitatively similar to that observed in the normal state of high-T_c superconductors.

Interacting one-dimensional (1D) electron systems generically exhibit *spin-charge separation*,[1] that is, the Hamiltonian separates at low energies into independent terms describing the charge and spin degrees of freedom. To date, the thermopower of interacting 1D electron systems has only been calculated for a few special cases, where the spin degrees of freedom reduce to noninteracting spins[2,3] or where the contribution of the spin excitations to the thermopower is negligible.[1] A more general result for the thermopower of such systems is of fundamental interest since the entropy carried by the spin excitations represents a qualitatively new type of thermopower, distinct from the familiar contributions of charge carrier diffusion, phonon drag, etc. Furthermore, Anderson has argued that the physics of the CuO_2 planes in high-T_c superconductors is that of spin-charge separation[4] and it is an interesting question whether their unusual normal-state thermopower[5-8] can be explained on that basis. Since a rigorous treatment of spin-charge separation in 2D systems is still lacking, it is clearly of interest to investigate the effects of spin-charge separation on the thermopower of 1D systems, for which powerful methods such as the Bethe ansatz are available.

In this letter, we calculate the contributions of both the charge and spin excitations to the low-temperature thermopower of the repulsive-interaction 1D Hubbard model in two limiting cases: in the strong-coupling limit and near the Mott-Hubbard metal-insulator transition occuring at half filling. In both of these limits, the charge degrees of freedom of the model can be mapped onto weakly interacting spinless fermions,[1,2,9,10] while the spin excitations can be shown to form an *ideal semion gas*[11] at low temperatures, which interacts with the charge degrees of freedom via a backflow condition that ensures that the electric current which flows in response to an electric field is really a current of *electrons*, which carry both charge and spin. The competition between the entropy carried by the charge excitations and that carried by the backflow of spin excitations leads to a nontrivial temperature and doping dependence of the thermopower. We comment briefly on the possible relevance of these results to an understanding of the normal-state thermopower of high-T_c superconductors and quasi-one-dimensional organic conductors.

The thermoelectric power S is given in the Kubo formalism by

$$S = \frac{1}{T}\left(\frac{\int_0^\infty \langle \hat{J}_E(0)\hat{J}_e(t)\rangle\, dt}{\int_0^\infty \langle \hat{J}_e(0)\hat{J}_e(t)\rangle\, dt} + \frac{\mu}{e}\right), \tag{1}$$

where $\hat{J}_e$ and $\hat{J}_E$ are the electric current and energy current operators in the Heisenberg representation, μ is the chemical potential, e is the absolute value of the electron charge, and $\langle\ \rangle$ denotes the thermal average. In a system with spin-charge separation, the energy current can be decomposed into a term associated with charge excitations and a term associated with spin excitations, $\hat{J}_E = \hat{J}_E^c + \hat{J}_E^s$, and the chemical potential can be written as $\mu = \mu_c + \mu_s$, where $\mu_c = \partial E_0/\partial N + \partial F_c/\partial N$ and $\mu_s = \partial F_s/\partial N$, E_0 being the ground-state energy, N the number of electrons in the system, and F_c and F_s the free energies of the charge and spin excitations. The thermopower can therefore be expressed as $S = S_c + S_s$, where S_c and S_s are defined by Eq. (1) with $\hat{J}_E^{c,s}$ and $\mu_{c,s}$ in place of $\hat{J}_E$ and μ. S_c and S_s can be interpreted as the entropies transported separately by charge and spin excitations when an electric current flows.

We specialize our arguments to the Hubbard model of spin-1/2 fermions hopping with matrix element t between nearest-neighbor sites of a 1D lattice with unit lattice constant and subject to a repulsive interaction U when two fermions (of opposite spin) occupy the same lattice site. Periodic boundary conditions are imposed on the lattice, which consists of L sites, and the system is threaded by a time-dependent magnetic flux $(\hbar c/e)\Phi(t)$.

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(In the following, we set $\hbar = 1$.) The complete excitation spectrum of this model was obtained by Woynarovich[12] for the case $\Phi(t) = 0$; we extend the results of Ref. 12 to $\Phi(t) \neq 0$ using the arguments of Ref. 13. In the large-L limit, the energy of the system with $N = L - H$ electrons can be expressed as[12]

$$E(L-H) = E_0(L) - \sum_{h=1}^{H} \varepsilon_c(k_h) + \sum_{\sigma=1}^{N_s} \varepsilon_s(\Lambda_\sigma), \quad (2)$$

where $E_0(L)$ is the ground-state energy at $N = L$, obtained in Ref. 14, $\varepsilon_c(k) = -2t\cos k -4t\int_0^\infty d\omega J_1(\omega)\cos(\omega \sin k)/[\omega + \omega\exp(\omega U/2t)]$, and $\varepsilon_s(\Lambda) = 2t\int_0^\infty d\omega J_1(\omega)\cos(\omega\Lambda)\,\mathrm{sech}(\omega U/4t)/\omega$. The momentum [defined mod(2π)] is given by[12]

$$P = \Phi(t) - \sum_{h=1}^{H} p_c(k_h) + \sum_{\sigma=1}^{N_s} p_s(\Lambda_\sigma) + \psi, \quad (3)$$

where $\psi = \pi(L - N/2 + N_s/2 + 1)$, $p_c(k) = k + 2\int_0^\infty d\omega J_0(\omega)\sin(\omega \sin k)/[\omega + \omega\exp(\omega U/2t)]$, and $p_s(\Lambda) = -\int_0^\infty d\omega J_0(\omega)\sin(\omega\Lambda)\,\mathrm{sech}(\omega U/4t)/\omega$. The parameters k_h and Λ_σ are real numbers satisfying $-\pi \leq k_h \leq \pi$, $-\infty \leq \Lambda_\sigma \leq \infty$, and can be interpreted as holes in the ground-state distributions of *pseudomomenta* and *spin rapidities* in the Lieb-Wu equations;[14] such excitations are referred to as *holons* and *spinons*. [In Eq. (2), we have omitted states with complex pseudomomenta for which there is a finite energy gap.[12]] The holons and spinons cannot in general be regarded as noninteracting quasiparticles since the k_h and Λ_σ are not free parameters but are determined by the asymptotic Bethe ansatz equations derived in Ref. 12:

$$\begin{aligned} Lp_c(k_h) = 2\pi I_h + \Phi(t) &+ \sum_{h'=1}^{H} \Theta_1(\sin k_h - \sin k_{h'}) \\ &+ \sum_{\sigma=1}^{N_s} \Theta_2(\sin k_h - \Lambda_\sigma) \\ &-\pi \sum_{\alpha=1}^{M_s} \mathrm{sgn}(\sin k_h - \mathrm{Re}\lambda_\alpha), \end{aligned} \quad (4)$$

$$\begin{aligned} Lp_s(\Lambda_\sigma) = -2\pi J_\sigma &- \sum_{h=1}^{H} \Theta_2(\Lambda_\sigma - \sin k_h) \\ &+ \sum_{\sigma'=1}^{N_s} \Theta_1(\Lambda_\sigma - \Lambda_{\sigma'}) - \sum_{\alpha=1}^{M_s} 2\tan^{-1}\frac{\Lambda_\sigma - \lambda_\alpha}{U/4t}, \end{aligned} \quad (5)$$

$$\sum_{\sigma=1}^{N_s} 2\tan^{-1}\frac{\lambda_\alpha - \Lambda_\sigma}{U/4t} = 2\pi K_\alpha + \sum_{\beta=1}^{M_s} 2\tan^{-1}\frac{\lambda_\alpha - \lambda_\beta}{U/2t}, \quad (6)$$

where M_s is the number of down-spin spinons and the two-body scattering phase shifts are given by $\Theta_1(x) = 2\int_0^\infty d\omega \sin(\omega x)/[\omega + \omega\exp(\omega U/2t)]$, $\Theta_2(x) = 2\tan^{-1}[\tanh(\pi t x/U)]$. The charge degrees of freedom are specified by the H quantum numbers I_h [defined mod(L)], which are distinct integers (half-integers) for M even (odd), M being the number of down-spin electrons. The spin degrees of freedom are specified by the N_s distinct quantum numbers J_σ, which take values in the range $-J_{\max}, -J_{\max}+1, \ldots, J_{\max}$, where $J_{\max} = (N - M - 1)/2$, and by the M_s quantum numbers K_α, which are integers (half-integers) for $N_s - M_s$ odd (even). The I_h and J_σ represent holes in the ground-state distributions of charge and spin quantum numbers in the Lieb-Wu equations, while the K_α and the complex parameters λ_α describe the stringlike states in the spin sector of the model (their relation to the parameters of the Lieb-Wu equations is given in Ref. 12). The z component of the total spin of the system is $S^z = (N-2M)/2 = (N_s - 2M_s)/2$. For the ground state in zero magnetic field, the I_h are consecutive integers (or half-integers) centered about $L/2$ and $N_s = N$ mod 2.

To evaluate the thermopower (1) using the above formalism would be quite difficult in general because, while the chemical potential can be extracted from the energy spectrum (2), the matrix elements of the current operators would have to be evaluated using the Bethe ansatz wave functions,[14,9] which are quite unwieldy. We therefore consider two limiting cases where the weakness of the holon-holon and spinon-spinon interactions allows one to construct the matrix elements of the current operators explicitly. Both for $H \ll L$ and for $U \gg t$, the holons can be mapped onto weakly interacting spinless fermions,[1,2,9,10] allowing the matrix elements of $\hat{J}_e$ and $\hat{J}_E^c$ to be evaluated;[15] in addition, for both of these cases one can show that in the low-temperature limit ($k_B T \ll v_s$) the energy spectrum of the spin excitations is

$$E_s \simeq v_s \sum_{\sigma=1}^{N_s} \left(\frac{\pi(1-\delta)}{2} - \left| \frac{2\pi J'_\sigma}{L} \right| \right) + \frac{\pi v_s N_s^2}{4L}, \quad (7)$$

where v_s is the spinon velocity in the low-energy limit, calculated in Refs. 16 and 1, $\delta \equiv |1 - N/L|$, and $J'_\sigma = J_\sigma + \mathrm{sgn}(J_\sigma)M_s/2$. Equation (7) follows upon linearizing the spinon dispersion relation in the low-energy ($\Lambda \to \pm\infty$) limit, replacing the spinon-spinon scattering phase shifts by their limiting low-energy form $\Theta_1(\Lambda_\sigma - \Lambda_{\sigma'}) \simeq (\pi/2)\mathrm{sgn}(\Lambda_\sigma - \Lambda_{\sigma'})$ and taking $\tan^{-1}[4t(\Lambda_\sigma - \lambda_\alpha)/U] \simeq (\pi/2)\mathrm{sgn}(\Lambda_\sigma)$. The low-energy spectrum (7) is equivalent to that of the "ideal spinon gas" described in Ref. 11: for fixed N_s, the degeneracies are equivalent to those of a system of spin-1/2 bosons, while the behavior of the Hilbert space as N_s is varied implies that the spinons are in fact *semions*, the second term in Eq. (7) representing the statistical interaction. The special feature of Haldane's ideal spinon gas is that the spinon-spinon interactions are described exactly by mean-field theory, so that spin-exchange processes between spinons are absent.[11] Such processes are implicit in Eqs. (5) and (6), which can be thought of as a nested Bethe-Yang ansatz for the spinons. However, spinon spin exchange processes have vanishing amplitude in the low-energy limit, so that the low-energy spectrum of the spin excitations (7) is independent of the quantum numbers K_α which specify the spin wave function of the spinons. The excitation spectrum (7) implies a low-temperature spin entropy per site of $\pi k_B^2 T/3v_s$,[11] from which it follows that

$$\lim_{T\to 0} \mu_s = \frac{\pi(k_B T)^2}{6v_s^2}\frac{\partial v_s}{\partial n}, \tag{8}$$

where $n \equiv N/L$. μ_s is not to be confused with the spinon chemical potential, which is zero since spinons are thermal excitations.

Because the spinon-spinon interactions have the mean-field form (7) in the low-energy limit, they do not contribute to the spinon energy current $\hat{J}_E^s$. Consequently, $\hat{J}_E^s$ commutes with the Hamiltonian in the low-energy limit and has the eigenvalue $J_E^s = v_s^2 P_s$ (assuming equal numbers of right and left movers), where P_s is the total momentum of the spinon gas. To calculate the contribution of the spinon energy current to the thermopower, we make use of the Onsager relation between the thermopower and the Peltier coefficient and define $\Pi^s \equiv TS_s = \Pi_E^s + \mu_s/e$, where $\Pi_E^s = \langle \hat{J}_E^s\rangle/\langle \hat{J}_e\rangle$ evaluated for a system with $\nabla T = 0$. Π_E^s depends on the details of the spinon relaxation processes; however, one can obtain an upper bound on Π_E^s by considering only the holon-spinon scattering. This upper bound will be adequate for our purposes since Π_E^s/T will prove to be negligible compared to the other contributions to the thermopower in the limits we consider. If we start with a system in thermal equilibrium and adiabatically generate an electric current $J_e = \langle \hat{J}_e\rangle$ (i.e., a shift in the k_h distribution) by applying an electric field $\mathcal{E} = -L^{-1}d\Phi/dt$,[15] the shift in the k_h distribution will lead via the spinon-holon scattering phase shifts $-\Theta_2(\Lambda_\sigma - \sin k_h)$ in Eq. (5) to a shift in the Λ_σ distribution. This backflow of spinons results because the electric field really couples to electrons, which carry both charge and spin. Both for $H \ll L$ and for $U \gg t$, one can approximate $\Theta_2(\Lambda_\sigma - \sin k_h) \simeq \Theta_2(\Lambda_\sigma) - \mathrm{sech}(2\pi t\Lambda_\sigma/U)\,(2\pi t \sin k_h/U)$; $\Theta_2(\Lambda_\sigma)$ is absorbed in a redefinition of the spinon momentum at finite H: $\tilde{p}_s(\Lambda_\sigma) = p_s(\Lambda_\sigma) + H\Theta_2(\Lambda_\sigma)/L$. Summing over $\tilde{p}_s(\Lambda_\sigma)$, we obtain

$$\langle \hat{J}_E^s\rangle = v_s^2 \left\langle \frac{1}{L}\sum_{\sigma=1}^{N_s} \frac{2\pi t}{U}\mathrm{sech}\frac{2\pi t\Lambda_\sigma}{U}\right\rangle_{J_e=0} \left\langle \sum_{h=1}^{H} \sin k_h\right\rangle_{J_e}, \tag{9}$$

which we evaluate explicitly below, using the fact that the sum over k_h is proportional to J_e, while the sum over Λ_σ is proportional to the excitation energy of the spinon gas, which can be evaluated via the correspondence with the model of Ref. 11. Equation (9) gives an upper bound to Π_E^s since spinon relaxation processes to the lattice have been neglected.

We first consider the limit $\delta \equiv |1-n| \ll 1$, close to the Mott-Hubbard metal-insulator transition. This case has been studied previously via a weak-coupling approximation;[1] we extend the results of Ref. 1 to arbitrary U, and explicitly verify the assertion of Ref. 1 that the contribution of the spin excitations to the thermopower is negligible in this limit. Equations (2) and (3) implicitly define an energy band $\varepsilon_c(k(p))$ for charge excitations, $k(p)$ being the inverse of the function $p_c(k)$. The holons can be thought of as holes in this energy band, which may be approximated near the energy minimum at $p = \pi$ by $\varepsilon_c(k(p)) \simeq \mu_- - (p-\pi)^2/2|m^*|$, where $\mu_- = \varepsilon_c(\pi)$ is the $T = 0$ chemical potential in the limit $n \to 1^-$,[14] and

$$|m^*| = \frac{1}{2t}\frac{\left\{1 - 2\int_0^\infty d\omega J_0(\omega)/[1+\exp(\omega U/2t)]^2\right\}}{1 - 2\int_0^\infty d\omega\, \omega J_1(\omega)/[1+\exp(\omega U/2t)]}$$

is the absolute value of the holon effective mass.[10,17] Equation (4) implies that the holon momenta $p_c(k_h)$ differ from those of noninteracting spinless fermions by a term which vanishes as H/L, $N_s/L \to 0$; we write $p_h \equiv p_c(k_h) = 2\pi I_h/L + \Phi/L + \delta p_h$, where δp_h has contributions from holon-holon, holon-spinon, and holon-string scattering. Holon-string scattering merely shifts the parity of the holon quantum numbers I_h and is therefore unimportant in the limit $L \to \infty$, while holon-holon and holon-spinon scattering give $\delta p_h/(p_h - \pi) \to -4\ln 2\,\delta/Up_c'(\pi) + CT^2$ as δ, $T \to 0$, where C is a U-dependent constant. The shifts of the holon momenta due to the interactions thus have a negligible effect on the charge excitation energies in the limit δ, $T \to 0$. Because of the vanishing interactions, the current operators $\hat{J}_e$ and $\hat{J}_E^c$ commute with the Hamiltonian[15] in the limit δ, $T \to 0$, and Eq. (1) can be evaluated straightforwardly to give

$$\lim_{\delta,T\to 0} S_c = \mathrm{sgn}(1-n)\,\frac{k_B^2 T}{3e}\frac{|m^*|}{\delta^2}, \tag{10}$$

where we have used the electron-hole symmetry of the model about $n = 1$.[12,10] The corrections to Eq. (10) due to holon-holon interactions are expected to be $\mathcal{O}(\delta^{-1})$. Evaluating Eqs. (8) and (9), we obtain for $\delta, T \to 0$, $\mu_s/eT = \mathrm{sgn}(n-1)\pi k_B^2 T/[12etI_1(2\pi t/U)]$, $|\Pi_E^s/T| < \pi^2 k_B^2 Tt|m^*|/[3eUp_c'(\pi)I_0(2\pi t/U)]$, where I_0 and I_1 are modified Bessel functions. S_s is thus negligible compared to S_c in the limit $\delta \to 0$. The low-temperature thermopower of the system thus becomes large and positive (hole-like) as the metal-insulator transition is approached from $n < 1$ and has the opposite sign (electronlike) as $n \to 1$ from above.

In the limit $U \gg t$, the full n dependence of the low-temperature thermopower can be calculated: When $U/t \to \infty$, $\varepsilon_c(k) \to -2t\cos k$, $p_c(k) \to k$, and the scattering phase shifts in Eq. (4) are $\mathcal{O}(t/U)$, leading to the well-known mapping of the holons onto noninteracting[15] spinless fermions in the strong-coupling limit.[2,9] The spinon dispersion relation now explicitly involves a contribution from the backflow of the holon distribution; Eqs. (2)–(6) give $v_s = (2\pi t^2/U)[1 - \sin(2\pi n)/2\pi n - 8\ln 2(t/\pi U)\sin^3 \pi n] + \mathcal{O}(t^4/U^3)$, which is consistent with the results of Refs. 16 and 1. The dominant contributions to the low-temperature thermopower come from S_c and μ_s/eT, which combine to give (for $n < 1$)

$$\lim_{\substack{U\to\infty\\T\to 0}} S = -\frac{k_B^2 T}{et}\left(\frac{\pi^2\cos\pi n}{6\sin^2\pi n}\right) - \frac{k_B^2 T}{3eJ}\left(\frac{\cos(2\pi n)/n - \sin(2\pi n)/2\pi n^2 + 24\ln 2\,(t/U)\sin^2\pi n\cos\pi n}{[1-\sin(2\pi n)/2\pi n - 8\ln 2\,(t/\pi U)\sin^3\pi n]^2}\right), \quad (11)$$

where $J = 4t^2/U$ is the antiferromagnetic superexchange constant and $S(2-n) = -S(n)$. The corrections to Eq. (11) are $\mathcal{O}(k_B^2T/eU)$ as $T\to 0$, and come from holon-holon interactions, higher-order terms in v_s, and from Eq. (9), which gives $|\Pi_E^s/T| < \pi^2k_B^2T/6eUn$. In the physically interesting parameter range $100 > U/t \gg 1$, Eq. (11) implies that the low-temperature thermopower is positive for all $n < 1$ and negative for all $n > 1$. The full temperature dependence of S_c [first term in Eq. (11)] can be calculated by the method of Ref. 2 (neglecting terms of order k_BT/U); as T increases, S_c monotonically approaches the high-temperature value $\mathrm{sgn}(1-n)(k_B/e)\ln[(1-\delta)/\delta]$.[3] The full temperature dependence of S_s [second term in Eq. (11)] is an open problem; however, when $k_BT \gg J$, μ_s is dominated by the spin entropy and $e\,\Pi_E^s/k_BT \ll 1$, so that $S_s \to \mathrm{sgn}(n-1)(k_B/e)\ln 2$, in agreement with the result of Ref. 2. Combining the high-temperature results for S_c and S_s yields the well-known Heikes formula.[3] For small hole dopings $\delta \ll 1$, S is dominated by S_c, which is large, positive, and a monotonic function of temperature. However, for $\delta > 1/3$, S is dominated by S_s, and is negative at high temperatures, but with a positive slope at low temperatures, implying the existence of a *positive peak* in the low-temperature thermopower. S has the opposite sign for electron doping.

The magnetic-field dependence of S is also readily obtained in the strong-coupling limit: a weak field B applied parallel to the chain does not couple to the charge degrees of freedom in the Peierls approximation, but the Zeeman coupling leads for $\mu B \ll k_BT \ll J$ to

$$\Delta S(B) = \frac{k_B}{e}\frac{k_BT}{\pi v_s}\frac{\partial \ln v_s}{\partial n}\left(\frac{\mu B}{k_BT}\right)^2. \quad (12)$$

It is interesting to compare the temperature and doping dependence of the thermopower in the 1D Hubbard model with that observed in the cuprate materials in which high-T_c superconductivity occurs, which are widely regarded to be quasi-two-dimensional doped Mott insulators.[4] The in-plane thermopower of the lightly doped cuprates is generically positive for hole doping[5] and negative for electron doping,[6,7] with a magnitude which increases drastically as the nominal concentration of doped carriers goes to zero, in qualitative agreement with Eq. (10). Upon further hole doping, the in-plane thermopower of the cuprates universally exhibits a positive peak at low temperatures, then decreases monotonically, often becoming negative at room temperature in the superconducting samples;[5] the mirror image behavior, with a negative peak at low temperatures, is exhibited[7] by the electron-doped superconductor $Nd_2CuO_{4-x}F_x$. Similarly, the unusual temperature dependence of the spinon backflow thermopower in the 1D Hubbard model leads for $\delta > 1/3$ and $U \gg t$ to a low-temperature peak in the thermopower which is positive for hole doping and negative for electron doping. Superconducting samples[6] of $Nd_{2-x}Ce_xCuO_4$ exhibit a peak of the same sign as that observed in the hole-doped cuprates, however, which cannot be accommodated in a model with electron-hole symmetry, such as the Hubbard model. The smallness of the isotropic contribution to the magnetothermopower[8,18] in high-T_c superconductors is qualitatively consistent with our result for the 1D Hubbard model, Eq. (12), which is reduced in magnitude by a factor of order k_BT/J compared to the high-temperature value.[19]

Our results should be of direct relevance for quasi-one-dimensional systems, such as organic conductors. The compound TTF[Ni(dmit)$_2$]$_2$, which can be modeled as a Hubbard chain with $n = 1/2$ and $U \gg t$ (so that the holon contribution to the thermopower is expected to be zero), has indeed been found[20] to exhibit a positive peak in the low-temperature thermopower and a large negative thermopower comparable to $-(k_B/e)\ln 2$ at high temperatures, as expected from spinon backflow.

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