Seeking large thermoelectric effects in MgO-based tunnel junctions

Xingtao Jia¹, Shizhuo Wang² and Minghui Qin³

¹ School of Physics and Electronic Information Engineering, Henan Polytechnic University, Jiaozuo 454000, People’s Republic of China
² Department of Physics, Beijing Normal University, Beijing 100875, People’s Republic of China
³ Institute for Advanced Materials and Laboratory of Quantum Engineering and Quantum Materials, South China Normal University, Guangzhou 510006, People’s Republic of China

E-mail: jiaxingtao@hpu.edu.cn

Abstract

There is much controversy concerning thermoelectric effects in the MgO-based magnetic tunnel junctions (MTJs) as reported in some recent publications. To clarify the problem, we give calculations from atomic first-principles systemically. Large Seebeck coefficient (S) and up-limit of figure of merit (ZT) are predicted in double- and multi-barrier MTJs, with those in single-barrier MTJs being relatively small. By restraining the phonon thermal conductance through the introduction of one vacuum barrier or numbers of MgO barriers, the up-limit of ZT can be obtained. Room temperature $S \approx -220 \mu V K^{-1}$ and $ZT \approx 3.5$ are predicted in an asymmetric Fe|MgO|Fe|Vacuum|Fe MTJs. The resonant quantum-well states are suggested to be responsible for the enhanced thermoelectric effects in the MTJs with double- and multi-barrier.

1. Introduction

Thermoelectric phenomena have recently been renovated by nanotechnology and spintronics [1–4]. Enhanced thermoelectric effects are found in nanostructures, which can be orders of magnitude larger than those in bulk systems. Therein, the localized states and phonon-scattering [5, 6] are considered to be responsible for the enhancement. One most used parameter for thermoelectric element is the dimensionless figure of merit $ZT = G S^2 T/\kappa$ with conductance $G$, Seebeck coefficient $S$, thermal conductance $\kappa$ at ambient temperature $T_0$. To seek large $ZT$, one should maximize $S$ and $G$, and minimize $\kappa$. There are two sources for $\kappa$: electron ($\kappa_e$) and phonon ($\kappa_p$). Both $\kappa_p$ and $\kappa_e$ can be engineered by modulating the structure parameters, and the up-limit of ZT can be obtained in the absence of $\kappa_p$.

Magnetic tunnel junctions (MTJs) show potential as the basic unit in heat-driving logic devices or memories [7]. Specifically, MgO-based MTJs are more attractive for huge tunnel magnetoresistance (TMR) [8] and tunnel magneto-Seebeck ratio (TMS) [7, 9–15]. Much controversies concerning thermoelectronic effects existed in MgO-based MTJs. Giant room temperature Seebeck coefficients range from 100 to 650 $\mu V K^{-1}$ have been reported in experiments [7, 9, 16, 17], which is about one order larger than theoretical [11] prediction of $\sim 10 \mu V K^{-1}$. Zhang et al. [18] argue that the huge Seebeck coefficient in experiments is related to the underestimation of temperature gradient during the measurements and simulations, and predict a room temperature $ZT \approx 10^{-2}$ in MgO-based MTJs. Thus, a detailed study on the thermoelectronic effects in MgO-based MTJs is still urgently needed.

There are two kinds of localized states contributing to the thermoelectric effects in MTJs: interface states and quantum well (QW) states. The latter has been well-studied in double-barrier MTJs (DBMTJs) and superlattice structures. QW states can greatly improve the thermoelectric effects in Bi$_2$Te$_3$ alloy-based superlattice structures [19]. Thermoelectric effects in ultrathin MgO-based MTJs is relatively small, where the free-electron-like tunneling states [8] around $\Gamma$ ($k_x = 0$, $k_y = 0$) point in the two-dimensional Brillouin zone (2D BZ) dominate electric and thermal transport. Resonant tunneling from the localized states [20–22] is the key to achieve large thermoelectric effects, which can be engineered by modulating the structure parameters such as barrier thickness and numbers, magnet type and thickness, defects concentration and position, and so on. If the
resonant states are shifted to the Fermi level ($E_F$), the energy dependence of the transmission would be strongly asymmetric, and the Seebeck coefficient would be large. In double- and multi-barrier MTJs, both interface states and QW states coexist \[23–25\], and the parameter space for thermoelectric effects is considerably larger than that in single-barrier MTJs. However, to the best of our knowledge, few works on this subject have been reported.

In this paper, we focus on the thermoelectric effects in the MgO-based MTJs in the presence of a nonequilibrium thermal distribution by applying the Landauer–Büttiker formalism with thermal bias \[26\]. At room temperature, we find $S \approx -220 \mu V K^{-1}$ and $ZT \approx 3.5$ in a junction with 7 atomic layer (L) MgO and 5 L vacuum barriers. The resonant QW states are considered to be responsible for the large thermoelectric effects.

This paper is organized as follows. In section 2, we give the details of our first-principle calculations. In section 3, we present our results on the MgO-based MTJs with symmetric and asymmetric structures. Section 4 is our summary.

2. Electronic structure and transport calculations

The electric voltage $\Delta V$ and thermal flow $Q$ in constrictions can be built by a current $I$ and temperature bias $\Delta T = T_R - T_L$ passing across the structure with relation: \[27, 28\]

$$\left( \frac{\Delta V}{Q} \right) = \left( \frac{1}{G} S \right) \left( \frac{I}{\kappa (\Delta T)} \right),$$

wherein the conductance $G$, Seebeck coefficient $S$ and electron thermal conductance $\kappa_e$ can be calculated from scatter matrix $S = [r \ t; t \ r]$ first-principle:

$$G = -G_0 \int dE T(E) \partial f / \partial E,$$

$$S = -\frac{G_0}{G} \frac{1}{T_0^2} \int dE (E - E_F) T(E) \partial f / \partial E,$$

$$\kappa_e = -G S^2 T_0 - \frac{G_0}{e^2 T_0} \int dE (E - E_F)^2 T(E) \partial f / \partial E,$$

with convolution of transmission $T(E) = Tr(\tau t^t)$, and kernel of $\partial f / \partial E$, $(E - E_F) \partial f / \partial E$ with Fermi–Dirac distribution functions $f(E) = \left[ e^{(E-E_F)/kT} + 1 \right]^{-1}$. Therein, $G_0 = 2e^2/h$ is quantum conductance, and $T_0 = (T_R + T_L)/2$ is ambient temperature. In figure 1, we give kernels used in the integrals above. When $T(E)$ varies slowly around $E_F$, $S = -e G_0 L_0 T_0 (\partial \ln T(E) / \partial E)|_{E_F}$ is reduced with Lorenz number $L_0 = 2.45 \times 10^{-8} V K^{-2}$.

Generally, phonon thermal conductance dominates over electron one, and the former can be greatly restrained with the order of $\sim 10^8 W K^{-1} m^{-2}$ by interfaces \[18, 29\]. When the lateral size of the thermoelectric element is shrunk to nanoscale, phonon thermal conductance decreases to one percent of that of the bulk as revealed in silicon nanowires by surface phonon scattering \[30, 31\]. Unfortunately, there are few data on the
relation between the phonon thermal conductance and lateral size of the MTJs. So, we introduce the up-limit of ZT, \( ZT_{(\text{max})} = GS^2T/\kappa_v \), to assess the potential of the thermoelectric elements.

In this study, we consider two-probe devices as Fe\(\mid\)MgO\(\mid\)Fe and Fe\(\mid\)MgO\(\mid\)Fe\(\mid\)MgO\(\mid\)Fe MTJs. We neglect small lattice mismatch at the Fe\(\mid\)MgO interface by fixing the interfacial atoms at their bulk positions, and assume the impurities existed at the first layer of MgO attached to Fe. In transport calculations, the electric current and thermal flow along the (001) direction, and a \(800 \times 800\) \(k\)-mesh is used to sample the 2D BZ to ensure excellent numerical convergence. More numerical details of the electronic structure and transport calculations can be found elsewhere [26, 32].

3. Thermoelectric effect in MgO-based MTJs

From energy-dependent transmission \( T(E) \), we can compute conductance \( G(T) \), electron thermal conductance \( \kappa_e \), and figure of merit ZT. At room temperature, \( G/S/\kappa_v \) are contributed mainly by energy range from \(-0.045/\pm0.015/\pm0.027\) eV to \(0.045/\pm0.085/\pm0.115\) eV with respect to \( E_F \), as estimated from the half-value width as shown in figure 1. So, ZT would be large as \( T(E) \) is strongly asymmetric around \( E_F \), and especially sharp peaks are existed within energy range \([E_F + 0.015\text{ eV}, E_F + 0.085\text{ eV}] \) or within the energy range \([E_F - 0.015\text{ eV}, E_F - 0.085\text{ eV}] \).

As \( T(E) \) in MgO-based MTJs is sensitive to the resonant states, one can engineer the shape of \( T(E) \) by modulating the structure parameters to achieve large thermoelectric effects. In the following, we study the effects of the structure parameters on the thermoelectric effects in the MgO-based MTJs systematically. Firstly, we take a look at single-barrier MTJs. Secondly, we turn to DBMTJs with symmetric and asymmetric structures. Thirdly, we study MTJs with multi-barriers. Finally, we pay attention to asymmetric DBMTJs with one MgO barrier and one vacuum barrier, where the up-limit of ZT would be achieved naturally in the absence of phonon thermal conductance.

3.1. Single-barrier MTJs

Figure 2 shows \( T(E) \) in ideal Fe\(\mid\)MgO\(\mid\)Fe MTJs with 3, 5 and 7 L MgO barriers. Generally, \( T(E) \) decreases exponentially as barrier thickness increases. In thick-barrier MTJs, \( T(E) \) is dominated by majority (\(\uparrow\)) spin around \(\Gamma\) point in 2D BZ, showing a free-electron-like tunnelling nature. When the barrier gets to 3 L thin, resonant tunneling from interfacial states in minority (\(\downarrow\)) spin (the hot spots in the insert of figure 2) begin to dominate the thermal transport, leading to larger positive \( S \sim 47 \mu V K^{-1} \) as shown in table 1 compared with small negative \( S \sim -12 \mu V K^{-1} \) in 7 L MgO MTJs. The Seebeck coefficient calculated here is consistent with theoretical work [11] but about one order smaller than that in the experiments [9, 16, 17] as mention above. One possible reason is the difference in band-match between experimental CoFe\(\mid\)MgO and calculated Fe\(\mid\)MgO MTJs, and another can be the temperature gradient measurements and simulations as discussed in a recent work [18]. The broaden peak in \( T(E) \) around \( E_F - 0.07\text{ eV} \) not only contributes to \( S \) but also to \( \kappa_v \), limiting room temperature ZT\(\text{(max)} \) to the order of \(10^{-1} \). Taking experimental phonon thermal conductance \( \kappa_p \sim 2.0 \times 10^8 \)
3.2. Double-barrier MTJs

The involvement of the QW states would change the thermoelectric effects in MgO-based DBMTJs. Firstly, we take a look at the thermoelectric transport in clean Fe $|$ MgO $|$ Fe MTJs, as shown in Figure 3. There are three striking peaks in $S$. The peak around $E_F - 0.07\,\text{eV}$ is from the resonant coupling between two interfacial states across the thin barrier, which follows the same position as that in Fe $|$ MgO $|$ Fe MTJs. The peaks around $E_F + 0.1\,\text{eV}$ and $E_F - 0.25\,\text{eV}$ are from resonant tunneling carried by QW states, which produce a bright transmission ring with peak value around one unit in 2D BZ as shown in the insert of Figure 3. As the barrier thickness increases, peaks in $S$ from the resonant QW states shift to left and decrease exponentially. At the same time, peaks from the resonant interfacial states shift less but decrease with ratio much faster than exponential form. As peaks in $S$ with energy below $E_F$ contribute to positive $S$, we observe peaks above $E_F$ dominate the thermoelectric effects. At room temperature, the DBMTJs with 3 L and 7 L MgO barriers show $S$ of $-46.5$ and $-210\,\mu\text{V K}^{-1}$, respectively, and the latter is almost completely from the resonant QW states.

By changing the thickness of the sandwiched Fe, the positions of the resonant peaks from the QW states in $S$ can be shifted to $E_F$, and larger thermoelectric effects may be achieved. In Table 2, we list the thermoelectric effects in DBMTJs with a series sandwiched Fe thickness. For MTJs with 3/7 L MgO barrier, the large room temperature $S \sim 80/200\,\mu\text{V K}^{-1}$ and up-limit of ZT $\sim 0.3/5.0$ is presented in MTJs with 30/16 L sandwiched Fe. Taking $\kappa_p \sim 2.0 \times 10^8\,\text{W K}^{-1}\text{m}^{-2}$ per barrier, room temperate ZT with order of $10^{-1}$ is observed in a DBMTJ with 3 L MgO barrier, which is about one order larger than that in single-barrier MTJs with the same barrier thickness.

In clean DBMTJs with ultrathin barriers, both the resonant QW and interfacial states would contribute to the thermoelectric effects. When an asymmetry is introduced into DBMTJs, the resonant interfacial states would be killed, while the resonant QW states are unaffected. For a clean Fe $|$ MgO $|$ Fe $|$ MgO $|$ Co DBMTJ, room temperature $S \sim -70\,\mu\text{V K}^{-1}$ and ZT with order of $10^{-1}$ is found in a junction with 3 L MgO barrier and 8 L sandwiched Fe.

### Table 1. Room temperature thermoelectric effect in ideal Fe $|$ MgO($m$) $|$ Fe SMTJs.

| $m$ (L) | $G$ ($10^{12}\,\Omega^{-1}\text{m}^{-1}$) | $S$ ($\mu\text{V K}^{-1}$) | $\kappa_p$ ($10^6\,\text{W m}^{-2}\text{K}^{-1}$) | ZT$^a$ | ZT$^b$ |
|---------|-----------------|----------------|-----------------|--------|--------|
| 3       | 17              | 47             | 113             | 0.035  | 0.098  |
| 5       | 0.75            | 15             | 6.3             | $0.27 \times 10^{-3}$ | $0.89 \times 10^{-3}$ |
| 7       | 0.071           | $-12$          | 0.53            | $0.14 \times 10^{-4}$ | $0.66 \times 10^{-3}$ |

$^a$ $\kappa_p \sim 2.0 \times 10^8\,\text{W K}^{-1}\text{m}^{-2}$ per barrier [18, 29].

$^b$ ZT(max).
3.3. multi-barrier MTJs

Although the Seebeck coefficient in MgO-based MTJs with single or double barriers is comparable to the well-studied thermoelectric materials, the optimistic room temperature ZT is around $10^{-1}$. The huge phonon thermal conductance would be responsible for the small ZT. By introducing more barriers (interfaces), the thermal conductance carried by phonon would be strongly restrained, and larger ZT is expected.

Figure 4(a) gives the $T(E)$ in the ideal MTJs with $2\sim7$ MgO barriers. Therein, the curves follow the same pattern in a large energy range, except small difference around $E_F+0.05\ eV$. A series of steps are present and the number of the steps increases as barriers number increases. The steps in $T(E)$ in multi-barrier MTJs is coming from the bright rings in 2D BZ as shown in the insert of figure 4(a), and a detailed analysis of their wave functions confirms that they are bonding-like and anti-bonding-like states resulting from the coupling between the QW states in the sandwiched Fe across the thin barrier. The small difference in the $T(E)$ curves in the MTJs with $2\sim7$ MgO barriers show less effect on the integrated thermoelectric effects such as $G$, $S$, $\kappa_v$, and up-limit of ZT, while the introducing of more MgO barriers have a large effect on the room temperature ZT as shown in

| $m$ | $l$ | $G$ (10$^2$ Ω$^{-1}$ m$^{-2}$) | $S$ (μV K$^{-1}$) | $\kappa_v$ (10$^6$ W m$^{-2}$ K$^{-1}$) | $ZT^a$ | $ZT^b$ |
|-----|-----|----------------|-----------------|----------------|--------|--------|
| 3   | 8   | $9.07(5.52)$   | $-4.9(-73)$     | $78.8(40.2)$   | $0.37 \times 10^{-4}(0.062)$ | $0.85 \times 10^{-3}(0.22)$ |
| 3   | 16  | $6.92(2.81)$   | $46.5(-54)$     | $65.3(31.9)$   | $0.027(0.018)$ | $0.069(0.076)$ |
| 3   | 24  | $7.62(3.42)$   | $80.1(26.4)$    | $61.2(41)$     | $0.091(0.51 \times 10^{-2})$ | $0.24(0.017)$ |
| 3   | 32  | $9.38$         | $80.2$          | $59.6$         | $0.11$ | $0.30$ |
| 5   | 8   | $1.08$         | $-79$           | $3.6$          | $0.019$ | $0.57$ |
| 5   | 12  | $0.494$        | $-135$          | $2.67$         | $0.026$ | $1.01$ |
| 5   | 16  | $0.305$        | $-119$          | $3.25$         | $0.011$ | $0.34$ |
| 5   | 20  | $0.25$         | $-67$           | $3.67$         | $0.33 \times 10^{-2}$ | $0.093$ |
| 7   | 8   | $0.139$        | $-68$           | $0.288$        | $0.19 \times 10^{-2}$ | $0.67$ |
| 7   | 12  | $0.0639$       | $-159$          | $0.109$        | $0.48 \times 10^{-2}$ | $4.44$ |
| 7   | 16  | $0.0318$       | $-210$          | $0.0803$       | $0.42 \times 10^{-2}$ | $5.23$ |
| 7   | 20  | $0.0203$       | $-209$          | $0.132$        | $0.26 \times 10^{-2}$ | $2.02$ |

$^a\ k_v \sim 2.1 \times 10^4$ W K$^{-1}$ m$^{-2}$ per barrier [18, 29].

$^b\ ZT$(max).

Figure 4. (a) Energy-dependent transmission and (b) temperature-dependent ZT in ideal MgO-based MTJs with 2–7 MgO barriers.

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Although the Seebeck coefficient in MgO-based MTJs with single or double barriers is comparable to the well-studied thermoelectric materials, the optimistic room temperature ZT is around $10^{-1}$. The huge phonon thermal conductance would be responsible for the small ZT. By introducing more barriers (interfaces), the thermal conductance carried by phonon would be strongly restrained, and larger ZT is expected.

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and in clean junction to 1 vac MgO 21 L 5L is obtained at and 1 OV in a Fe kMgO interface, the peaks in MgO fi
Although a up-limit of ZT in the junction are more marked at low temperatures. The room temperature ZT almost increases linearly with the increase of the barrier number, leading to ZT 1 when barrier numbers are larger than 400. In the calculations, we suppose that the junction is infinite in lateral size, allowing the calculations simple but a little different from the real setups. As the phonon thermal conductance is sensitive to the size of lateral lattice, especially for the nanoscale devices, a MTJs with barrier numbers much less than 400 maybe possible to achieve ZT 1 in the experiment.

3.4. Asymmetric Fe | MgO | Fe | Vac | Fe DBMTJs
Although a up-limit of ZT 5 is found in a DBMTJ with 7 L MgO barrier as list in table 2, the real ZT is relatively small for the huge phonon thermal conductance. One simple way to restrain is introducing a vacuum barrier [33]. Substituting one MgO barrier by vacuum in the DBMTJ, the up-limit of ZT would be achieved by blocking phonon thermal constance.

Figure 5(a) shows the T (E) in ideal Fe | MgO | Fe(24) | Vac | Fe DBMTJs. The introduction of the vacuum changes both the position and magnitude of the tunneling peaks in T (E) from the resonant QW states, and almost quenches the peaks from the resonant interfacial states, as comparing with that in the clean Fe | MgO | Fe | MgO | Fe DBMTJs as shown in figure 3(a). The MTJs with thicker barrier show sharper peaks in T (E), leading to larger S and ZT in thicker barrier MTJs than those in the thinner one. Both S and ZT deviate from linear pattern as temperature increases, as shown in figures 5(b) and (c). The saturation S 320 µV K 1 is obtained at 100 K and ZT 5.8 at 175 K in a junction with 7 L MgO and 5 L vacuum barriers. Thus, thermoelectric effects in the junction are more marked at low temperatures.

Similar to symmetric Fe | MgO | Fe | MgO | Fe DBMTJs, the thermoelectric effects in Fe | MgO | Fe | Vac | Fe can be scissored by the thickness of the sandwiched Fe, as listed in table 3. Room temperature S 45/172 µV K 1 and ZT 0.1/3.5 are achieved in a junction with 3/7 L MgO and 3/5 L vacuum barriers and 24 L sandwiched Fe. Two factors are responsible to the sharper ZT in thicker barrier junction than that in the thinner one, one is the larger S and another is the larger electronic conductance to thermal conductance ratio G/κe. Both factors are related to the sharper peak in T (E) in thick barrier junction, comparing with thinner one. From the evolvement of the shape of T (E) in Fe | MgO | Fe | Vac | Fe DBMTJs with respect to the barrier thickness as shown in figure 5(a), it is reasonable to deduce that the peaks in T (E) would be even sharper in junctions with MgO barrier thickness more than 7 L, and even larger room temperature thermoelectric effects are expected in such junction.

The most prevailing imperfection in MgO-based MTJs is oxygen vacancy (OV). When OV exists at the Fe|MgO interface, the peaks in T(E) are broadened and shift compared to clean junctions, leading to an enhanced κe, and weakened S, G, and ZT. For example, 5% OV in a Fe | MgO(7) | Fe(16) | MgO(7) | Fe DBMTJ would deduce room temperature S and G from 210 µV K 1 and 3.18 1019 Ω 1 m 2 in clean junction to 147 µV K 1 and 1.12 1019 Ω 1 m 2, respectively. At the same time, the electron thermal conductance κe...
increase from $0.08 \times 10^6 \text{ W m}^{-2} \text{ K}^{-1}$ to $0.29 \times 10^6 \text{ W m}^{-2} \text{ K}^{-1}$, leading to a reduction of 95% in ZT. So, to achieve large thermoelectricity, the MTJs should be as clean as possible.

The nature of enhanced thermoelectric effects in the MgO-based MTJs with double- and multi-barrier is the involvement of resonant QW states. Insulator | metal | insulator structure is the basis of QW states, and materials in the MTJs open a new parameter space for thermoelectric effects. So, enhanced thermoelectric effects in MTJs with double- and multi-barrier can be a general phenomena.

So far, we have discussed the effect of a group of structure parameters on the thermoelectric effects in the MgO-based MTJs, and give an example to achieve the up-limit of ZT. Besides the structure parameters discussed above, exterior parameters can also be used to modulate the thermoelectric effects such as texture engineering [34], electrons (holes) doping [35, 36], electric bias [7, 37, 38], strain and stress [39, 40], and so on. From the up-limit of ZT predicted first-principlely, one can see there are much space to improve the thermoelectric effects in the MgO-based MTJs.

4. Summary

Based on atomic first principles, we compute the thermoelectric effects in the MgO-based MTJs. The effects of the structure parameters on the thermoelectric effects are discussed systematically such as barrier thickness, barrier numbers, sandwiched Fe thickness, asymmetric barrier, and so on. We predict the large Seebeck coefficient and up-limit of ZT in double- and multi-barrier MTJs. In a multi-barrier MTJs, the linearly increased ZT with respect to the number of MgO barriers is predicted, where Seebeck coefficient is constant while thermal conductance (mainly from phonon) decreases linearly. By restraining the phonon thermal conductance by vacuum barrier, the up-limit of ZT can be obtained. At room temperature, Seebeck coefficient $S \approx -220 \text{ \mu V \ K}^{-1}$ and ZT $\approx 3.5$ are predicted in an asymmetric Fe $|$ MgO(7)$|$ Fe(24)$|$ Vacuum(5)$|$ Fe MTJs. The resonant QW states near Fermi energy are considered to be responsible for the enhanced thermoelectric effects in the double- and multi-barrier MTJs.

Acknowledgments

XJ thanks supports from K Xia at BNU, and we gratefully acknowledge financial support from National Natural Science Foundation of China (11274094 and 51332007). XJ also acknowledge financial support from Henan Polytechnic University (B2012-021 and T2016-2).

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| $m$ (L) | $n$ (L) | $l$ (L) | $G$ (10$^2$Ω$^{-1}$m$^{-2}$) | $S$ ($\mu$V/K) | $\kappa_v$ ($10^4$W/m$^2$K) | ZT |
|---|---|---|---|---|---|---|
| 3 | 3 | 8 | 15.4 | 15 | 94.0 | 0.010 |
| 3 | 3 | 16 | 8.9 | −45 | 53.3 | 0.102 |
| 3 | 3 | 24 | 5.67 | −30 | 55.9 | 0.028 |
| 3 | 3 | 32 | 5.84 | 20 | 60.0 | 0.012 |
| 5 | 4 | 8 | 1.01 | 55 | 5.91 | 0.154 |
| 5 | 4 | 16 | 0.895 | −68 | 2.27 | 0.540 |
| 5 | 4 | 24 | 0.338 | −144 | 1.77 | 1.19 |
| 5 | 4 | 32 | 0.227 | −72 | 3.51 | 0.101 |
| 7 | 5 | 8 | 0.0589 | 78.6 | 0.312 | 0.35 |
| 7 | 5 | 16 | 0.0657 | −67.8 | 0.127 | 0.71 |
| 7 | 5 | 24 | 0.0247 | −172 | 0.0628 | 3.50 |
| 7 | 5 | 32 | 0.0115 | −144 | 0.156 | 0.459 |
