How bilayer excitons can greatly enhance thermoelectric efficiency

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Abstract
Currently, one of the major nanotechnological challenges is to design thermoelectric devices that have a high figure of merit. To that end, we propose to use bilayer excitons. Bilayer exciton systems are shown to have an improved thermopower and an enhanced electric counterflow and thermal conductivity, with respect to regular semiconductor-based thermoelectrics. Here we present a roadmap towards experimental realization of a bilayer exciton thermocouple. A bilayer exciton heterostructures of $p$- and $n$-doped Bi$_2$Te$_3$ can have a figure of merit $zT \sim 60$. Another material suggestion is to make a bilayer out of electron-doped SrTiO$_3$ and hole-doped Ca$_3$Co$_4$O$_9$.

Ranging from household refrigeration to waste heat in power plants, the problem of converting heat into electricity or vice versa is of paramount technological importance. Instead of solving this issue with compression-based heat pumps and the like, solid state materials might offer a resolution. Among the notable advantages of using solid state Peltier or Seebeck devices are their light weight, the absence of any moving parts, and the elimination of need for any environmentally unfriendly substances. However, the main problem is to devise a thermoelectric device with sufficient efficiency.

The efficiency of a solid-state thermoelectric material is commonly expressed in terms
Figure 1: **Traditional thermocouple.** A traditional thermocouple device consists of a $n$-type material (in green) and a $p$-type material (in red) connected electrically in series, but parallel with respect to the temperature gradient. The charge carriers move from the hot end to the cold end (black arrows), generating an electrical current (white dashed arrows).

of the dimensionless *figure of merit*[^1]

$$zT = \frac{\alpha^2 / L}{1 + \frac{\kappa_{ph}}{\kappa_e}}$$

where $\alpha_e$, $\kappa_{ph}$, $\kappa_e$ and $L$ are the Seebeck coefficient, the phonon thermal conductivity, the thermal conductivity of the electrons and the Lorenz number, respectively. Following the Wiedemann-Franz law, the Lorenz number $L$ is defined by[^2]

$$L = \frac{\kappa_e}{\sigma_e T},$$

where $\sigma_e$ is the electrical conductivity and $T$ is the temperature. For most semiconductors and metals, the Lorenz number turns out to be universal and given by $L_{WF} = \frac{\pi^2}{3} \left( \frac{k_B}{e} \right)^2 = 2.44 \times 10^{-8} \, \text{V}^2 \, \text{K}^{-2}$.

So how does one proceed to maximize this figure of merit? Obviously by increasing the thermopower $\alpha$ and decreasing the phonon thermal conductivity $\kappa_{ph}$. Indeed, the central
The maxim of modern thermoelectric research is summarized by the phrase “phonon glass - electron crystal” [3]. That implies using doped heavy element semiconductors with carrier concentrations \( n \sim 10^{18} - 10^{20} \text{ cm}^{-3} \) [4]. Furthermore, specific low-dimensional nanostructures can be used to suppress the phonon contribution to the thermal conductivity. An example of a “state-of-the-art” thermoelectric material is the hole-doped semiconductor \( \text{Bi}_2\text{Te}_3 \). It has a large Seebeck coefficient of \( \alpha = 220 \mu \text{V K}^{-1} \) at room temperature and can be constructed such that the phonon thermal conductivity is low, \( \kappa_{\text{ph}} \approx \kappa_e \). This yields a figure of merit of \( zT \approx 1 \) [5]. Unfortunately, a \( zT \) of about 1 is not serious competition to conventional heat pumps regarding efficiency. Instead, we need to find new materials to enhance \( zT \) to 4 or greater [1]. In this Article we present a concept that can lead potentially to an enhancement of the figure of merit \( zT \) by more than a factor of 40. For this we need to mobilize bilayer excitons.

An exciton is the bound state of an electron and a hole. In a bulk material the electron and hole can recombine and thus annihilate the exciton. In order to avoid this annihilation, it has been proposed to spatially separate the electrons and holes in two different layers [6, 7]. Such a heterostructure of a \( p \)-type and \( n \)-type material can support bilayer excitons. Since the bound state of an electron and a hole is effectively a boson, it can Bose condense, and substantial experimental and theoretical efforts have been devoted to the realization of a bilayer exciton condensate [8, 9, 10, 11, 12, 13].

The bilayer exciton system displays a remarkable similarity to a thermoelectric couple, as shown in Fig. 1. Such a device consists of a \( p \)-type and \( n \)-type thermoelectric material connected in series [1]. A heat gradient will cause the charge carriers to diffuse from the hot side to the cold side, thus converting the heat gradient into an electrical current. Now imagine that one brings the \( p \)- and \( n \)-type material close to each other. At distances of order nanometers the Coulomb attraction between the electrons and holes will bind them...
Figure 2: **Bilayer exciton thermocouple.** In a bilayer exciton thermocouple, as presented in this paper, the $p$- and $n$-type materials are brought close together so that bilayer excitons can form. In the thermoelectric device, the excitons move from the hot side to the cold side (orange arrows) generating an electric counterflow (white dashed arrows). Due to the bosonic nature of the excitons, the exciton thermocouple can have a higher figure of merit than the traditional thermocouple of Fig. 1.

into bilayer excitons. As shown in Fig. 2 these excitons will flow from the hot side to the cold side. Even though excitons are charge neutral, the fact that the holes and electrons are spatially separated implies that an exciton current amounts to two opposite currents in the two layers.[14] If one closes the circuit in the counterflow set-up, see Fig. 2, the exciton current is converted into an electrical current.

Though most bilayer exciton research focusses on the condensation of excitons, for thermoelectric applications uncondensed excitons are required. Although the counterflow conductivity of the exciton Bose condensate is infinite due to the superfluidity, the Seebeck coefficient vanishes faster and the figure of merit drops to zero.[15] However, we will argue here that the high-temperature properties of a gas of bilayer excitons will lead to an exceptionally large thermopower, combined with a small Lorenz number. In addition,
Table 1: Room temperature properties of traditional thermoelectrics compared to bilayer exciton systems. Here we compare three traditional thermoelectrics (doped Bi$_2$Te$_3$[1, 5], doped SrTiO$_3$[34, 35, 36] and Ca$_3$Co$_4$O$_9$[34, 35, 36]) with two bilayer exciton thermocouples. The Bi-based exciton bilayer consists of $p$- and $n$-doped Bi$_2$Te$_3$, the oxide-based exciton bilayer is a SrTiO$_3$/Ca$_3$Co$_4$O$_9$ heterostructure. The columns display the optimal carrier density $n$/(cm$^{-3}$), degeneracies $g_s g_0$, thermopower $\alpha$($\mu$VK$^{-1}$), phonon thermal conductivity $\kappa_{ph}$ (W m$^{-1}$K$^{-1}$), electron/exciton thermal conductivity $\kappa_e (\kappa_{ex})$ (W m$^{-1}$K$^{-1}$) and the figure of merit $zT$. For bilayer excitons, the $\kappa_{ph}$ is taking into account the contribution of phonons in both layers. Clearly, the exciton systems exhibit a tremendous enhancement of the thermoelectric figure of merit $zT$.

| materials                             | $n$     | $g_s g_0$ | $\alpha$ | $\kappa_{ph}$ | $\kappa_e (\kappa_{ex})$ | $zT$ |
|---------------------------------------|---------|-----------|-----------|----------------|--------------------------|------|
| doped Bi$_2$Te$_3$                    | $10^{19}$ | 2 x 6     | 220       | 0.8            | 0.8                      | 1.0  |
| Bi-based exciton bilayer              | $3 \times 10^{18}$ | $4 \times 36$ | 1060      | 1.6            | 7.0                      | 63   |
| Nb-doped SrTiO$_3$                    | $6 \times 10^{20}$ | 2 x 3     | 150       | 8.2            | 0.9                      | 0.08 |
| Ca$_3$Co$_4$O$_9$                     | $1.9 \times 10^{21}$ | 2         | 120       | 3.0            | 0.5                      | 0.08 |
| Oxide-based exciton bilayer           | $1.0 \times 10^{21}$ | 4 x 3     | 353       | 11.2           | 8.3                      | 3.6  |

the remarkably weak coupling between excitons and phonons, the dominant scattering mechanism at room temperature, gives rise to a high exciton thermal which reduces the effect of the phonon thermal conductivity $\kappa_{ph}$.

These are the two pillars of bilayer exciton thermoelectrics: better thermopower, and better conductivities. We introduce two possible material choices for the bilayer exciton thermocouple: a Bi-based system of $p$- and $n$-doped Bi$_2$Te$_3$, and an oxide system consisting of $n$-doped SrTiO$_3$ and $p$-type Ca$_3$Co$_4$O$_9$. The enhancement of thermopower and thermal conductivities are shown in Table 1, which comprises our main result. In the remainder of this Letter we will describe both pillars in detail, ending with a roadmap towards experimental realization of the bilayer exciton thermocouple.

**Enhanced thermopower**

The most relevant factor that determines the figure of merit is the thermopower. For Fermi liquid materials such as metals or degenerate semiconductors, the thermopower
Seebeck coefficient $\alpha$ is given by \[^2\]

$$|\alpha| = \frac{\pi^2 k_B}{3} e \left( \frac{T}{T_F} \right),$$

(3)

where $k_B$ is the Boltzmann constant, $e$ is the elementary electric charge and $T_F$ is the Fermi temperature of the material. Therefore, a natural way to increase the thermopower is to reduce the charge carrier density $n$, since $T_F \sim n^{2/3}$.

Bilayer excitons, on the other hand, behave like hard-core bosons on a lattice, and for temperatures well above the condensation temperature $T_c$ and in the dilute limit, these particles behave as classical particles. In that case the thermopower is given by the entropy per particle\[^{16, 17}\]. The entropy is given by the logarithm of the number of possible configurations, whereby spin degeneracy $g_s$ and orbital degeneracy $g_o$ play an important role.\[^{18, 19}\] For excitons on a crystalline lattice with density $\rho$ per unit cell, we find that the excitonic thermopower is given by

$$\alpha_{\text{ex}} = \frac{k_B}{e} \left( \ln g_s g_o + \ln \frac{1 - \rho}{\rho} \right).$$

(4)

The exciton spin degeneracy is usually $g_s = 2 \times 2 = 4$ since both the electron and hole can have two spin states, and the singlet-triplet splitting is negligible compared to the temperature. Similarly, the exciton orbital degeneracy $g_o$ is the product of the electron and hole orbital degeneracy.

In Table 1 we compare the Seebeck coefficient of traditional thermoelectric materials to bilayer exciton systems. It is clear that the orbital and spin degeneracies give rise to a large thermopower for the bilayer exciton thermocouple.

Recall that for most free electron system, the Lorenz number was found to be constant\[^2\]. However, in the case of excitons, we no longer have free electrons but instead we are dealing with bosons. In the counterflow construction of Fig. 2 excitons are transported as if
they have a charge $e$ and a mass of $2m_e$. At high temperatures, the Lorenz number for dilute bosons of charge $e$ is given by

$$L_{\text{ex}} = 2 \left( \frac{k_B}{e} \right)^2,$$

which is smaller than the Wiedemann-Franz value by a factor $L_{\text{WF}} / L_{\text{ex}} \approx 1.6$.

**Enhanced conductivity**

Now we discuss the thermal conductivity of excitons. It is known that bulk excitons have an exceptionally large mobility. For example, in cuprous oxide Cu$_2$O at low temperatures the exciton mobility is found to be $\mu_{\text{ex}} \sim 10^7$ cm$^2$/Vs$^2$[21], orders of magnitude larger than the mobility of charged carriers in this compound[22]. Measurements on low-temperature bilayer exciton mobility in GaAs/AlGaAs double quantum wells show similarly high mobilities[23]. Recent results at $T = 100$ K in GaAs/AlGaAs double quantum wells suggest that the high mobility of excitons persists up to higher temperatures[24].

Following the sparse experimental results, we can make a qualitative prediction for the room temperature exciton counterflow conductivity. For any kind of particle, its resistance depends on impurity scattering, scattering with phonons and particle-particle scattering. In a pure dilute sample at room temperatures, the dominant contribution to the resistivity is given by phonon-scattering.

In the case of charged carriers, the scattering with acoustic phonons depends on the deformation potential $D$. Since excitons are charge-neutral, interaction with lattice ions is naturally much weaker. The leading order long-wavelength matrix element between excitons and acoustic phonons is given by[8, 25]

$$\sqrt{\frac{2\hbar k}{M u}} (D_e - D_h)$$

(6)
where \( k \) is the phonon wavevector, \( u \) is the speed of sound, \( M \) is the ion mass, and \( D_{e,h} \) are the deformation potentials of the electron and hole, respectively.\[^{8, 25}\] The minus sign in Eqn. (6) is due to the opposite charge of the electron and hole, and leads to the relatively small exciton-phonon coupling. This cancellation depends on the size of the exciton: the radius of a bulk exciton in Cu_2O is approximately 7 Å, hence as long as the interlayer excitons are of similar size we expect the above theory to hold.

Thus, if one is able to construct a heterostructure such that the deformation potentials \( D_{e,h} \) for the electrons and holes are almost equal, one finds an extremely low exciton-phonon scattering rate. In fact, relative to the electron-phonon scattering rate,

\[
\frac{\tau_{\text{ex-ph}}}{\tau_{\text{e-ph}}} \sim \left( \frac{D}{\Delta D} \right)^2
\]

where \( \Delta D \) is the absolute difference between the electron deformation potential and the hole deformation potential. From the scattering rate \( \tau \), the electric conductivity is given by \( \sigma = ne^2\tau/m \). The exciton mass is approximately twice the electron mass, \( m_{\text{ex}} \approx 2m_e \). Using the Lorenz number we then find the change in thermal conductivity of the excitons with respect to the electronic thermal conductivity,

\[
\frac{\kappa_{\text{ex}}}{\kappa_e} = \frac{\tau_{\text{ex-ph}}}{\tau_{\text{e-ph}}} \frac{m_e}{m_{\text{ex}}} \frac{L_{\text{ex}}}{L_{\text{WF}}} \sim 0.3 \times \left( \frac{D}{\Delta D} \right)^2 .
\]

Now imagine that one is able to create a 10% difference between the deformation potentials of the electrons and holes. This will lead to a factor fifty better electric conductivity, and a factor thirty better thermal conductivity of the excitons. We consider this estimate, which we used to obtain the results of Table 1, reasonable given the experimental results quoted earlier.
Conclusions: a roadmap

The idealized exciton thermocouple could therefore have a $zT$ of more than 60, see Table 1. This is, however, the optimistic theorist perspective. In reality, many pitfalls and engineering problems will lower the figure of merit. Nonetheless, with the ideal $zT \sim 60$ there is much room for shortfalls to still arrive at a sizable figure of merit.

The biggest experimental challenge is to actually make bilayer excitons without condensing them, just like it is difficult to get uncondensed electron-electron pairs. Most material proposals for bilayers exciton condensation, such as semiconductor quantum wells, double layer graphene or topological insulators function in the BCS limit, which means that bilayer excitons are not bound at temperatures above the condensation temperature. Instead, we need materials with strong exciton binding, which can be achieved by bringing the layers close to each other (of the order of several unit cells), to minimize the electron and hole kinetic energy and to ensure that the electrons and holes have similar effective mass and deformation potential. For this end we propose two possible material choices for the bilayer exciton thermocouple: bismuth-based bilayers and oxide-based bilayers.

Oxide heterostructures, that themselves show good thermoelectric properties, will show a tremendous enhancement of the figure of merit. Many oxide materials have a layered lattice structure, such as $n$-doped SrTiO$_3$, and the $p$-type cobaltates $\text{Ca}_3\text{Co}_4\text{O}_9$ and $\text{Na}_x\text{CoO}_2$. Techniques such as molecular beam epitaxy (MBE) and pulsed laser deposition (PLD) make it currently possible to grow heterostructures of layered oxide materials with unit-cell precision. We propose to construct a bilayer system with a few unit cell layers of Nb-doped SrTiO$_3$, then about 6 unit cell layers of an oxide insulator like SrTiO$_3$ to avoid recombination of electrons and holes, and
Figure 3: **Density dependence of the thermoelectric properties.** a. Here we show the figure of merit $zT$, the thermopower $\alpha$, the phonon thermal conductivity $\kappa_{\text{ph}}$ and the exciton thermal conductivity $\kappa_{\text{ex}}$ for the $p$- and $n$-doped Bi$_2$Te$_3$ bilayer. The optimal value of $zT$ is used in Table 1. b. Same as Fig. a, but for the bilayer of $n$-doped SrTiO$_3$ and $p$-type Ca$_3$Co$_4$O$_9$.

On top of that a few unit cell layers of the $p$-type Ca$_3$Co$_4$O$_9$. This construction should be repeated in a superlattice structure, with insulating materials in between each bilayer, to obtain a material with a bulk thermoelectric effect. Using ion-beam etching ramp-edges can be made, so that one can separately contact each $n$-type and $p$-type layer using for example deposition of gold contacts.

We have chosen these materials because they can be fabricated with similar charge carrier densities of $n \sim 10^{20}$ cm$^{-3}$, and the carriers have similar room temperature electronic thermal conductivities $\kappa_e \sim 0.5$ W m$^{-1}$ K$^{-1}$ [37, 39, 40]. The SrTiO$_3$ have 3-fold orbital degeneracy, the cobaltate is a one-band material. Together with the spin entropy, we expect that at the given densities the exciton thermopower is $\alpha_{\text{ex}} \sim 650$ $\mu$V K$^{-1}$, which is even a conservative estimate given the $\alpha \sim 1000$ $\mu$V K$^{-1}$ results in SrTiO$_3$ superlattices [38]. Given our earlier analysis we expect that the exciton thermal conductivity is greatly enhanced. Since the phonon thermal conductivity in oxides is usually the limiting factor in obtaining a high $zT$, the large exciton thermal conductivity yields
several orders of magnitude enhancement of $zT$, see Table [1]. The density-dependence of $\alpha$, $\kappa_{\text{ex}}$ and $zT$ for the oxides is displayed in Fig. [3a], showing that for a wide window of exciton densities the SrTiO$_3$/Ca$_3$Co$_4$O$_9$ bilayer system has a sizable figure of merit.

The other possibility is to form heterostructures of $p$- and $n$-doped Bi$_2$Te$_3$ [5], separated at a distance of $\sim$ 2-3 nm. Molecular beam epitaxy (MBE) has been used extensively to fabricate nanostructures of bismuthtellurides. The fact that we have the same material used for both the $p$-type and $n$-type layer implies that the exciton binding and the exciton-phonon coupling are optimal. Together with the sixfold band degeneracy of Bi$_2$Te$_3$ we find an extraordinary large Seebeck coefficient, $\alpha \sim 1115 \, \mu V \, K^{-1}$. We thus find an extremely large $zT \sim 70$, see Table [1]. The density-dependence of the thermopower and the thermal conductivity is shown in Fig. [3b].

The main question is of course whether excitons would form at room temperature. If we take the insulating layer to have a dielectric constant of about $\epsilon \sim 20$, then at a distance of 2.3 nm the electron-hole attraction is $V \sim 30 \, \text{meV} = 350 \, \text{K}$. Given this estimate of the binding energy, most of the electrons and holes will be bound into bilayer excitons at room temperature.

Nonetheless, at room temperature it is likely that bilayer excitons coexist with unbound electrons and holes. Naturally, this will reduce the figure of merit, as some of the currents are carried by the less-efficient electrons and holes. A further experimental challenge is therefore to dilute the number of unbound electrons and holes, for example by bringing the $p$ and $n$-type materials closer to each other or to change the insulator layer.

In conclusion, we have shown qualitatively that the thermoelectric efficiency of a bilayer exciton system can be significantly enhanced. The nanotechnological engineering of thermoelectric devices should therefore aim at using bilayer excitons, instead of electrons
and holes, to generate electricity out of heat.

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