Spin Heat Accumulation Induced by Tunneling from a Ferromagnet

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SPIN-WIEDEMANN-FRANZ LAW

Here we develop in more detail the concept of a spin-Wiedemann-Franz law used in the main text. First, we prove mathematically the aforementioned law for the case of a spin relaxation dominated by elastic spin-flip scattering. Next, we include inelastic relaxation processes and define the parameter \( \alpha \). Finally, we show that for elastic spin-flip scattering a non-zero spin heat accumulation does not induce nor change the spin accumulation.

Spin heat resistance and Wiedemann-Franz law

The standard way to compute the steady state value of the spin accumulation \( \Delta \mu \) is to balance the net amount of spins injected per unit time \( t \), with the loss of spins due to spin relaxation in the non-magnetic material. The latter is governed by the spin resistance \( r_s \), for which \( r_s = \rho \lambda_s \) is obtained if we assume that \( \Delta \mu \) decays exponentially away from the tunnel interface with a spin-relaxation length \( \lambda_s \) \[1–3\]. In a similar way we evaluate the spin heat resistance \( r_s^Q \), which describes how effective a (non-equilibrium) spin heat accumulation \( \Delta T_s \) is relaxed to zero in a non-magnetic material. Inelastic (electron-phonon and electron-electron) interactions are possible relaxation mechanisms. We shall include these later on, but we first discuss another mechanism, namely, elastic (or quasi-elastic) spin-flip scattering. The net effect is that it moves electrons above the Fermi energy from the hot spin reservoir to the cold spin reservoir, and simultaneously moves electrons below the Fermi energy in the opposite direction. This results in cooling of the hot spin reservoir and warming up of the colder spin reservoir, thus equalizing \( T^\uparrow \) and \( T^\downarrow \). Note that there is no net flow of spin angular momentum if the spin-flip scattering rate is not dependent on energy (i.e., unlike a spin accumulation, a non-zero \( \Delta T_s \) does not give rise to spin relaxation, even though spin-flip scattering is the relevant scattering process. See the last subsection for an explicit evaluation).

The energy flow between the two spin reservoirs in the non-magnetic material is denoted by the spin heat current \( J_s^{QV} \) per unit volume (in \( \text{Wm}^{-3} \)). We also introduce \( \kappa_s^V \), the volume spin heat conductance (in \( \text{Wm}^{-2} \text{K}^{-1} \)) that connects the two spin reservoirs, such that

\[ J_s^{QV} = \kappa_s^V \Delta T_s \]  \hspace{1cm} (15)

The total energy flow is obtained by integrating \( J_s^{QV} \) over the full spatial extent of the spin heat accumulation, noting that according to heat diffusion, the spin heat accumulation is expected to decay exponentially with vertical distance \( z \) from the injection interface. That is, \( \Delta T_s(z) = \Delta T_s \exp(-z/\lambda_Q) \), where \( \lambda_Q \) is the spin heat relaxation length. In a steady state, the integrated \( J_s^{QV} \) has to be equal to the spin heat current \( J_s^Q \) that is injected through the tunnel interface. Thus

\[ I_s^Q = \int_0^\infty J_s^{QV} \, dz = \kappa_s^V \int_0^\infty \Delta T_s(z) \, dz \]
\[ = \kappa_s^V \Delta T_s \lambda_Q \]  \hspace{1cm} (16)

Comparing this to the relation that defines \( r_s^Q \) (Eq. (8) in the main text) we find that

\[ r_s^Q = \frac{1}{2 \kappa_s^V \lambda_Q} \]  \hspace{1cm} (17)

Below it is shown that if the spin heat accumulation relaxes via elastic or quasi-elastic spin-flip scattering (only), we have for \( \kappa_s^V \)

\[ \kappa_s^V = \frac{L_0 T_0}{2 \rho (\hbar c)^2} = \frac{L_0 T_0}{2 r_{el}^s \lambda_s^2} \]  \hspace{1cm} (18)

Because in that case also \( \lambda_Q = \lambda_s^{el} = \lambda_s^{el} \), we finally obtain

\[ r_s^Q = \frac{r_{el}^s}{L_0 T_0} \]  \hspace{1cm} (19)

This is an important result, as it constitutes a type of spin-Wiedemann-Franz law, relating the electronic spin resistance \( r_s \) to the spin heat resistance \( r_s^Q \) via the Lorentz number and the temperature.

The final task is to prove Eq. (18) for \( \kappa_s^V \). We assume that the spin-flip scattering is predominantly elastic. The spectral (energy-resolved) spin heat current due to spin-flip scattering, \( J_s^{QV}(\varepsilon) \), in units of \( \text{Wm}^{-3} \text{eV}^{-1} \), is then

\[ J_s^{QV}(\varepsilon) = \frac{2 \varepsilon N(\varepsilon) \left[ f(\varepsilon, \mu^+, T^\uparrow) - f(\varepsilon, \mu^+, T^\downarrow) \right]}{r_{el}^s(\varepsilon)} \]  \hspace{1cm} (20)
where \( \epsilon \) is the energy with respect to the Fermi energy, \( f(\epsilon, \mu, T) \) is the Fermi-Dirac distribution function, \( N(\epsilon) \) is the density of states per spin in the non-magnetic material, and \( \tau_{sf}^{\text{el}} \) is the spin-flip time. The factor of 2 appears because each spin flip reduces the energy difference between the two spin reservoirs by two units of \( \epsilon \). Note that \( \tau_{sf}^{\text{el}} \) and the elastic spin-relaxation time \( \tau_{sf}^{\text{el}} \) are related by \( \tau_{sf}^{\text{el}} = 2 \tau_{sf}^{\text{el}} \). If the density of states and the spin-flip time are not strongly dependent on energy, the spin heat current per unit volume is given by

\[
J_s^{QV} = \int_{-\infty}^{\infty} J_s^{QV}(\epsilon) \, d\epsilon
\]

\[
= \frac{N(\epsilon_F)}{\tau_{sf}^{\text{el}}} \int_{-\infty}^{\infty} \left[ f(\epsilon, \mu^+, T^+) - f(\epsilon, \mu^-, T^+) \right] \epsilon \, d\epsilon
\]

The following relations are of use:

\[
f_i = f_0 + \frac{\partial f_0}{\partial \mu} (\mu_i - \mu_0) + \frac{\partial f_0}{\partial T} (T_1 - T_0)
\]

\[
\frac{\partial f_0}{\partial \mu} = - \frac{\partial f_0}{\partial \epsilon}
\]

\[
\frac{\partial f_0}{\partial T} = - \frac{\partial f_0}{\partial \epsilon} \frac{\epsilon}{T_0}
\]

where \( f_0(\epsilon, \mu_0, T_0) \) is the equilibrium distribution parameterized by \( \mu_0 \) and \( T_0 \). Using these relations we have

\[
f(\epsilon, \mu^+, T^+) - f(\epsilon, \mu^-, T^+) = - \frac{\partial f_0}{\partial \mu} \Delta \mu - \frac{\partial f_0}{\partial \epsilon} \frac{\epsilon^2}{T_0} \Delta T_s
\]

Inserting this into Eq. (21) gives

\[
J_s^{QV} = \frac{N(\epsilon_F)}{\tau_{sf}^{\text{el}}} \int_{-\infty}^{\infty} \left[ - \frac{\partial f_0}{\partial \epsilon} \Delta \mu - \frac{\partial f_0}{\partial \epsilon} \frac{\epsilon^2}{T_0} \Delta T_s \right] \epsilon \, d\epsilon
\]

The integral can be evaluated using the Sommerfeld expansion (see Eq. (3) of Hatami et al. Ref. 4):

\[
\int_{-\infty}^{\infty} \left( - \frac{\partial f_0}{\partial \epsilon} \right) \epsilon \, d\epsilon = 0
\]

\[
\int_{-\infty}^{\infty} \left( - \frac{\partial f_0}{\partial \epsilon} \right) \epsilon^2 \, d\epsilon = \frac{\pi^2}{3} (kT_0)^2 = L_0 T_0^2 e^2
\]

We then obtain

\[
J_s^{QV} = \frac{N(\epsilon_F)}{\tau_{sf}^{\text{el}}} L_0 T_0 \Delta T_s
\]

Next we use the generalized Einstein relation, \( \mu_e n^{\text{tot}} = D_e \frac{\partial n_{\text{tot}}}{\partial E_F} \), where \( \mu_e \) is the carrier mobility, \( D \) the diffusion constant, \( n_{\text{tot}} \) is the spin-integrated electron density, and \( \frac{\partial n_{\text{tot}}}{\partial E_F} \) is equivalent to \( 2 N(\epsilon_F) \) (recall that the latter was defined per spin). Using the resistivity \( \rho = 1/(n_{\text{tot}} e \mu_e) \), \( \lambda_s = \sqrt{D \tau_{sf}^{\text{el}}} \), and \( \tau_{sf}^{\text{el}} = \rho \lambda_s \), we obtain

\[
J_s^{QV} = \frac{1}{2 \rho (\lambda_s^2)} L_0 T_0 \Delta T_s = \frac{L_0 T_0}{2 \tau_{sf}^{\text{el}} \lambda_s^2} \Delta T_s
\]

Comparing this to Eq. (15) gives

\[
\kappa_s^V = \frac{L_0 T_0}{2 \tau_{sf}^{\text{el}} \lambda_s^2}
\]

just as it was already used in Eq. (18). Note that the spin heat current due to spin-flip scattering is proportional solely to \( \Delta T_s \). There is no contribution from the non-zero \( \Delta \mu \). This is a direct consequence of the assumption that the spin-flip time is not dependent on energy.

**Wiedemann-Franz law including inelastic relaxation**

In the previous subsection we derived a Wiedemann-Franz type of law for spin resistance, under the assumption that the relaxation of the spin heat accumulation occurs exclusively via (quasi-) elastic spin-flip scattering. However, spin heat relaxation occurs also via inelastic scattering processes.

One inelastic process is electron-phonon (e-ph) scattering. Phonons in the non-magnetic material cause an indirect energy flow from the hot spin reservoir to the cold spin reservoir, which can be understood as follows. In the presence of a non-zero \( \Delta T_s \), the temperature difference \( \Delta T_{\text{e-ph}} \) between electrons and phonons is spin dependent and given by \( T^+ - T^0 \) and \( T^+ - T^0 \), respectively. As a result, the heat transfer to the phonons is spin dependent, which tends to equalize \( T^+ \) and \( T^0 \). For the specific case where the phonon temperature is equal to the spin-averaged electron temperature \( T_0 \), we have \( \Delta T_{\text{e-ph}} = +\Delta T_s/2 \) and \( \Delta T_{\text{e-ph}} = -\Delta T_s/2 \).

Another process is inelastic electron-electron (e-e) scattering, which causes a direct energy flow between the two spin reservoirs in the non-magnetic material, which also tends to equalize \( T^+ \) and \( T^0 \). Note that in the absence of spin-orbit scattering [5] the e-e interaction does not cause any additional relaxation of the spin accumulation \( \Delta \mu \), so its sole effect is to decrease \( \Delta T_s \).

Including relaxation via inelastic processes in the derivation of the spin-Wiedemann-Franz law has important effects. First, in Eq. (15) for the spin heat current \( J_s^{QV} \) induced by a non-zero \( \Delta T_s \), we must add the volume spin heat conductances \( \kappa_v^{V,e-ph} \) and \( \kappa_v^{V,e-e} \) due to e-ph and e-e interactions, respectively. Therefore,

\[
J_s^{QV} = (\kappa_s^V + \kappa_v^{V,e-ph} + \kappa_v^{V,e-e}) \Delta T_s
\]

where we have kept \( \kappa_v^V \) to denote the term due to elastic spin-flip scattering, given by Eq. (31).

The second effect is that we can no longer set \( \lambda_s = \lambda_Q \), as previously done in Eq. (17) for the spin heat resistance. Let us discuss this in more detail. In the regime of diffusive (heat) transport, we expect that \( \lambda_Q = \sqrt{D \tau_Q} \), with the corresponding diffusion constant being the same as that for charge transport. This is a valid assumption since electronic heat transport is associated with electrons only. This is analogous to the case of electronic transport.
spin transport where the charge and spin diffusion coefficients coincide. On the other hand, the inelastic scattering processes contribute to the relaxation of the spin heat accumulation, and therefore affect \( \tau_Q \). This spin heat relaxation time can be obtained by defining the inelastic contribution \( \tau_Q^{\text{inel}} \) to it via

\[
\kappa_s^{V,e-ph} + \kappa_s^{V,e-e} = \frac{N(\varepsilon_F) e^2 L_0 T_0}{\tau_Q^{\text{inel}}} (33)
\]
such that, together with \( \kappa_s^V = N(\varepsilon_F) e^2 L_0 T_0/\tau_Q^{el} \) from Eq. (29), we have the total volume spin heat conductance

\[
\kappa_s^{V} + \kappa_s^{V,e-ph} + \kappa_s^{V,e-e} = \frac{N(\varepsilon_F) e^2 L_0 T_0}{\tau_Q} (34)
\]

where

\[
\tau_Q = \tau_s^{el} \left( \frac{\kappa_s^V}{\kappa_s^{V,el} + \kappa_s^{V,e-ph} + \kappa_s^{V,e-e}} \right) (36)
\]

For the case of \( \kappa_s^V \gg \kappa_s^{V,e-ph} + \kappa_s^{V,e-e} \), this reduces to \( \tau_Q^{el} \). Using Eq. (29) we can now write

\[
\tau_Q = \tau_s \left( \frac{\kappa_s^V}{\kappa_s^{V,el} + \kappa_s^{V,e-ph} + \kappa_s^{V,e-e}} \right) (36)
\]

The spin heat resistance including inelastic relaxation processes is obtained after Eq. (17) as

\[
\tau_s^{Q} = \frac{1}{2 \left( \kappa_s^{V,el} + \kappa_s^{V,e-ph} + \kappa_s^{V,e-e} \right) \lambda_Q} \frac{\tau_Q}{\tau_s} = \frac{\tau_Q}{\tau_s} \left( \frac{\kappa_s^{V,el} + \kappa_s^{V,e-ph} + \kappa_s^{V,e-e}}{\kappa_s^{V,el}} \right) \lambda_Q (38)
\]

where we have first inserted Eq. (36), then used Eq. (31) to eliminate \( \kappa_s^V \) and then used that \( \tau_Q^{el} = (\lambda_Q/\lambda_s^{el})^2 \).

Finally, expressing the spin resistance \( r_s^{Q} \) due to elastic processes only in terms of the total spin resistance \( r_s = \rho \lambda_s^{el} \) including inelastic processes, we finally obtain

\[
r_s^{Q} = \frac{r_s}{L_0 T_0} \times \frac{1}{\alpha} (39)
\]

which is the spin-Wiedemann-Franz law as described in Eq. (13) of the main text.

**Spin relaxation with finite spin heat accumulation**

For the sake of completeness, we evaluate the spin current due to elastic spin-flip scattering in the presence of a spin heat accumulation using the same approach as in the first subsection. It is demonstrated that a non-zero \( \Delta T_s \) does not induce nor change the spin accumulation, provided that the spin-flip time (and the density of states around the Fermi level) are not dependent on energy in the relevant range of a few \( kT \). The spectral spin current density per unit volume (in Am\(^{-3}\)) is

\[
J_s^V(\varepsilon) = \frac{2 N(\varepsilon) \left[ f(\varepsilon, \mu^+, T^+) - f(\varepsilon, \mu^-, T^-) \right]}{\tau_Q^{el}(\varepsilon)} e (40)
\]

If \( \tau_Q^{el} \) and \( N \) do not vary much around the Fermi energy, the integrated spin current in the presence of a non-zero \( \Delta \mu \) and a non-zero \( \Delta T_s \) is

\[
J_s^V = \frac{N(\varepsilon_F) e}{\tau_Q^{el}} \int_{-\infty}^{\infty} \left[ f(\varepsilon, \mu^+, T^+) - f(\varepsilon, \mu^-, T^-) \right] d\varepsilon (41)
\]

Using Eq. (25) as in the first subsection, this is rewritten as

\[
J_s^V = \frac{N(\varepsilon_F) e}{\tau_Q^{el}} \int_{-\infty}^{\infty} \left[ \frac{\partial f_0}{\partial \varepsilon} \Delta \mu + \frac{\partial f_0}{\partial \mu} \frac{\varepsilon}{T_0} \Delta T_s \right] d\varepsilon (42)
\]

The second part of the integral, which contains \( \Delta T_s \) vanishes (see Eq. (3) of Hatami et al. Ref. 4). Thus, only a non-zero \( \Delta \mu \) produces a net spin current and spin relaxation. Now, using

\[
\int_{-\infty}^{\infty} \left( \frac{\partial f_0}{\partial \varepsilon} \right) d\varepsilon = 1 (43)
\]

and

\[
\frac{N(\varepsilon_F) e}{\tau_Q^{el}} = \frac{1}{2 e \rho (\lambda_s^{el})^2} (44)
\]

we obtain

\[
J_s^V = \frac{1}{2 e \rho (\lambda_s^{el})^2} \Delta \mu (45)
\]

Assuming that \( \Delta \mu(z) = \Delta \mu \exp(-z/\lambda_s) \), and using that in a steady state, the spatially integrated \( J_s^V \) has to be equal to the spin current \( I_s \) that is injected through the tunnel interface, we obtain

\[
I_s = \int_{0}^{\infty} J_s^V dz = \frac{1}{2 e \rho (\lambda_s^{el})^2} \int_{0}^{\infty} \Delta \mu(z) dz
\]

\[
= \frac{1}{2 e \rho (\lambda_s^{el})^2} \Delta \mu = \frac{1}{2 e r_s^{el}} \Delta \mu (46)
\]

where \( r_s^{el} = \rho \lambda_s^{el} \) is the spin resistance (in Ohm\(^2\)), consistent with the definition of \( r_s \) in Eq. (7) of the main text.
HANLE EFFECT AND BLOCH EQUATIONS

In this section we describe the dynamics of the spin accumulation and the spin heat accumulation due to the Hanle effect. A spin accumulation in a paramagnetic material implies that there is a net non-equilibrium magnetization or particle spin density \( N_s \). The particle spin density is linearly proportional to the spin accumulation \( \Delta \mu \) if the latter is sufficiently small:

\[
N_s = (N^\uparrow - N^\downarrow) \frac{\hbar}{2} = \frac{\hbar}{2} \int_{-\infty}^{\infty} N(\varepsilon) \left[ f(\varepsilon, \mu^\uparrow, T_n^0) - f(\varepsilon, \mu^\downarrow, T_n^0) \right] d\varepsilon \\
= N(\varepsilon_F) \frac{\hbar}{2} \int_{-\infty}^{\infty} \left[ \left( -\frac{\partial f_0}{\partial \varepsilon} \right) \Delta \mu + \left( -\frac{\partial f_0}{\partial \varepsilon} \right) \frac{\varepsilon}{T} \Delta T_s \right] d\varepsilon \\
= N(\varepsilon_F) \frac{\hbar}{2} \Delta \mu
\]  

(47)

Similarly, if we denote the energy density by \( E \), then the energy spin density \( E_s = E^\uparrow - E^\downarrow \) is proportional to the spin heat accumulation \( \Delta T_s \):

\[
E_s = E^\uparrow - E^\downarrow = \int_{-\infty}^{\infty} N(\varepsilon) \left[ f(\varepsilon, \mu^\uparrow, T_n^0) - f(\varepsilon, \mu^\downarrow, T_n^0) \right] \varepsilon d\varepsilon \\
= N(\varepsilon_F) \int_{-\infty}^{\infty} \left[ \left( -\frac{\partial f_0}{\partial \varepsilon} \right) \varepsilon \Delta \mu + \left( -\frac{\partial f_0}{\partial \varepsilon} \right) \frac{\varepsilon^2}{T} \Delta T_s \right] d\varepsilon \\
= N(\varepsilon_F) \varepsilon^2 L_0 T_0 \Delta T_s
\]  

(48)

When a magnetic field is applied transverse to the initial spin direction (Hanle geometry), the electron magnetic moments precess at the Larmor frequency. In the next paragraph we first describe the resulting dynamics of the particle spin density using the Bloch equations, and then apply a similar analysis to the energy spin density to describe the dynamics of the spin heat accumulation.

The Bloch equations that describe the dynamics (precession and relaxation) of the particle spin density are:

\[
\frac{\partial N_{s,x}}{\partial t} = \gamma (N_s \times B)_x - \frac{N_{s,x}}{\tau_s} \\
\frac{\partial N_{s,y}}{\partial t} = \gamma (N_s \times B)_y - \frac{N_{s,y}}{\tau_s} \\
\frac{\partial N_{s,z}}{\partial t} = \gamma (N_s \times B)_z - \frac{N_{s,z}}{\tau_s}
\]  

(49-51)

with \( \gamma = g \mu_B / \hbar \) the gyromagnetic ratio. For \( B \) applied along the z-axis, and the boundary conditions \( N_{s,x} = N_0 \) and \( N_{s,y} = N_{s,z} = 0 \) at \( t = 0 \), the solutions are:

\[
N_{s,x} = N_0 \cos(\omega_L t) e^{-t/\tau_s} \\
N_{s,y} = N_0 \sin(\omega_L t) e^{-t/\tau_s} \\
N_{s,z} = 0
\]  

(52-54)

Thus, the precession of the magnetic moments in a transverse magnetic field causes the projection of the particle spin density onto the x and y axis to oscillate at the Larmor frequency, whereas spin relaxation causes an exponential decay. Since the particle spin density has a magnitude and a direction that varies in time, and the particle spin density is proportional to the spin accumulation, the latter also has a magnitude and direction that varies in time. Integrating over time gives a Lorentzian for the x-component of the particle spin density, and similarly for the x-component of the spin accumulation:

\[
N_{s,x} \propto \langle \Delta \mu \rangle_x = \langle \Delta \mu \rangle_0 \frac{1}{1 + (\omega_L \tau_s)^2}
\]  

(55)

The energy spin density \( E_s = E^\uparrow - E^\downarrow \) describes the difference in the energy of electrons with spin pointing parallel (\( \uparrow \)) and antiparallel (\( \downarrow \)) to a given quantization axis. The precession of the electron magnetic moments in a transverse magnetic field causes the quantization axis to be time dependent, and hence the energy spin density also precesses at the Larmor frequency. To describe this, we introduce a similar set of Bloch equations, but now for the energy spin density, and taking into account that a different relaxation time \( \tau_Q \) should be used:

\[
\frac{\partial E_{s,x}}{\partial t} = \gamma (E_s \times B)_x - \frac{E_{s,x}}{\tau_Q} \\
\frac{\partial E_{s,y}}{\partial t} = \gamma (E_s \times B)_y - \frac{E_{s,y}}{\tau_Q} \\
\frac{\partial E_{s,z}}{\partial t} = \gamma (E_s \times B)_z - \frac{E_{s,z}}{\tau_Q}
\]  

(56-58)

Please note that the energy spin density can be defined even if the particle spin density is zero. For \( B \) applied along the z-axis, and with similar boundary conditions \( E_{s,x} = E_0 \) and \( E_{s,y} = E_{s,z} = 0 \) at \( t = 0 \), the solutions are thus similar to Eqs. (52), (53) and (54), namely:

\[
E_{s,x} = E_0 \cos(\omega_L t) e^{-t/\tau_Q} \\
E_{s,y} = E_0 \sin(\omega_L t) e^{-t/\tau_Q} \\
E_{s,z} = 0
\]  

(59-61)

Since the energy spin density has a magnitude and a direction that varies in time, and it is proportional to the spin heat accumulation, the latter also has a magnitude and direction that varies in time, effectively making the spin heat accumulation a (time-dependent) vector. Integrating over time gives a Lorentzian for the x-component:

\[
\langle \Delta T_s \rangle_x = \langle \Delta T_s \rangle_0 \frac{1}{1 + (\omega_L \tau_Q)^2}
\]  

(62)

In the main text it was shown that the voltage across the tunnel contact has two independent contributions from, respectively, the spin accumulation and the spin heat accumulation. This applies to the static case as well as under dynamic conditions. The Hanle voltage signal is thus a superposition of two voltage signals:

\[
\Delta V_{Hanle} = \left( \frac{P_G}{2e} \right) \Delta \mu + \left( \frac{P_L S}{2} \right) \Delta T_s
\]  

(63)
The Hanle line shape is thus also a superposition of two Lorentzian curves with different amplitude and line width, determined by $\Delta \mu$ and $\Delta T_s$ and by $\tau_s$ and $\tau_Q$:

$$
\Delta V_{\text{Hanle}}(B_z) = \left( \frac{P_L}{2e} \right) \frac{(\Delta \mu)_0}{1 + (\omega_L \tau_s)^2} + \left( \frac{P_L S}{2} \right) \frac{(\Delta T_s)_0}{1 + (\omega_L \tau_Q)^2}
$$

(64)

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