Dispersion of active particles of arbitrary shape

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3D Standard ABP model

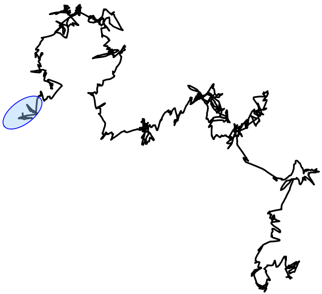
Stochastic equations for the 3D active Brownian particle (ABP) model:

$$\dot{\boldsymbol{x}} = U\,\boldsymbol{e} + \left\{\sqrt{2D_\parallel}\,\mathbb{P}^\parallel(\boldsymbol{e}) + \sqrt{2D_\perp}\,\mathbb{P}^\perp(\boldsymbol{e})\right\}\cdot\dot{\boldsymbol{w}}_1$$
$$\dot{\boldsymbol{e}} = -2D_\mathrm{r}\,\boldsymbol{e} + \sqrt{2D_\mathrm{r}}\,\boldsymbol{e}\times\dot{\boldsymbol{w}}_2\,.$$

- translational noises $D_\perp$ and $D_\parallel$ along and perpendicular to the direction of swimming $\boldsymbol{e}$;
- the rotational noise D_r affects the swimming direction;
- diffusivities are related to particle mobility $\times k_B T$;
- $\boldsymbol{w}_i(t)$ are independent standard Wiener processes (5 total);
- angular drift $-2D_\mathrm{r}\,\boldsymbol{e}$ ensures unit length $\boldsymbol{e}$.

[Peruani & Morelli (2007); van Teeffelen & Löwen (2008); Baskaran & Marchetti (2008); Romanczuk & Schimansky-Geier (2011); Romanczuk *et al.* (2012); Kurzthaler *et al.* (2016); Kurzthaler & Franosch (2017); Ai *et al.* (2013); Solon *et al.* (2015); Zöttl & Stark (2016); Wagner *et al.* (2017); Redner *et al.* (2013); Stenhammar *et al.* (2014); Chen & Thiffeault (2021)]

3D Standard ABP model: Effective diffusivity



An ABP meanders around, and for long times there is a well-known formula for its effective diffusivity:

$$\boxed{D_{\text{eff}} = \|\boldsymbol{U}\|^2 / 6 D_{\text{r}}}$$

This is a very useful dispersion result that can be measured experimentally.

The ABP model is not quite general: only one vector $\boldsymbol{e}$ is used to denote the orientation. This is fine if the particle has hydrodynamic axial symmetry.

Goal: investigate the general particle and derive D_{eff}.

Secondary goal: avoid using Euler angles or similar parametrization!

General 3D body

For an arbitrary body, instead of $\boldsymbol{e}$ we use an orthogonal matrix $\mathbb{Q}$.

In general, need 6 coordinates: $\boldsymbol{x} = (x_1, x_2, x_3)$ and $\boldsymbol{\phi} = (\phi_1, \phi_2, \phi_3)$.

- $\boldsymbol{\phi}$ is some vector of coordinates for $\mathrm{SO}(3)$ specifying the particle orientation as $\mathbb{Q}(\boldsymbol{\phi})$ (*e.g.*, Euler angles, quaternions).
- Notation: A hat $\widehat{\ }$ will indicate a matrix or vector with 6 rows and/or columns, denoting position and angles:

$$\text{e.g.,} \quad \widehat{\boldsymbol{x}} = (\boldsymbol{x}, \boldsymbol{\phi})$$

Force-velocity relation

In Stokes flow, force $\boldsymbol{f}$ and torque $\boldsymbol{\tau}$ are linearly related to the particle's velocity $\boldsymbol{u}$ and angular velocity $\boldsymbol{\omega}$:

$$\begin{pmatrix} \boldsymbol{u} \\ \boldsymbol{\omega} \end{pmatrix} = \widehat{\mathbb{M}} \cdot \begin{pmatrix} \boldsymbol{f} \\ \boldsymbol{\tau} \end{pmatrix} \qquad \widehat{\mathbb{M}} := \begin{pmatrix} \mathbb{M}_{\boldsymbol{x}\boldsymbol{x}} & \mathbb{M}_{\boldsymbol{x}\boldsymbol{\phi}} \\ \mathbb{M}_{\boldsymbol{\phi}\boldsymbol{x}} & \mathbb{M}_{\boldsymbol{\phi}\boldsymbol{\phi}} \end{pmatrix}.$$

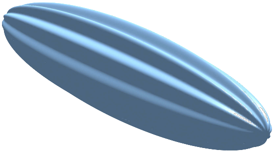
- The 6×6 symmetric matrix $\widehat{\mathbb{M}}$ is called the grand mobility matrix.
- $\widehat{\mathbb{M}}$ is symmetric, so $\mathbb{M}_{\boldsymbol{x}\boldsymbol{\phi}} = \mathbb{M}_{\boldsymbol{\phi}\boldsymbol{x}}^\top$.
- Torque (and thus $\widehat{\mathbb{M}}$) is defined w.r.t. a reference point.

The coupling matrix

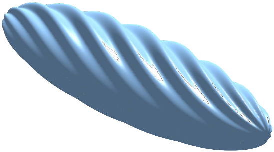
$$\widehat{\mathbb{M}} = \begin{pmatrix} \mathbb{M}_{\boldsymbol{xx}} & \mathbb{M}_{\boldsymbol{x\phi}} \\ \mathbb{M}_{\boldsymbol{\phi x}} & \mathbb{M}_{\boldsymbol{\phi\phi}} \end{pmatrix}.$$

- There is a unique reference point called the center of hydrodynamic reaction for which $\mathbb{M}_{\boldsymbol{x\phi}} = \mathbb{M}_{\boldsymbol{x\phi}}^\top$ [*e.g.*, Happel & Brenner (1983)].
- If the center of mass differs from the center of reaction, then $\mathbb{M}_{\boldsymbol{x\phi}}$ cannot be symmetric.
- In particular, it cannot be zero.
- Let's call such a particle wobbly: $\mathbb{M}_{\boldsymbol{x\phi}} \neq \mathbb{M}_{\boldsymbol{x\phi}}^\top$.
- Even a sphere with nonuniform density is wobbly.

Hydrodynamic chirality



$\mathbb{M}_{\boldsymbol{x}\boldsymbol{\phi}} = 0$



$\mathbb{M}_{\boldsymbol{x}\boldsymbol{\phi}} \neq 0$

$$\widehat{\mathbb{M}} = \begin{pmatrix} \mathbb{M}_{\boldsymbol{x}\boldsymbol{x}} & \mathbb{M}_{\boldsymbol{x}\boldsymbol{\phi}} \\ \mathbb{M}_{\boldsymbol{\phi}\boldsymbol{x}} & \mathbb{M}_{\boldsymbol{\phi}\boldsymbol{\phi}} \end{pmatrix}.$$

- If the coupling $\mathbb{M}_{\boldsymbol{x}\boldsymbol{\phi}}$ is nonzero for any choice of reference point, a particle is hydrodynamically chiral.
- This can coincide with geometric chirality, as above.

Overdamped stochastic dynamics

Kinematics:

$$\dot{\boldsymbol{x}} = \boldsymbol{u}, \qquad \dot{\boldsymbol{\phi}} = \mathbb{L} \cdot \boldsymbol{\omega} \,.$$

The tensor $\mathbb{L}$ depends on the specific coordinate representation of $\mathrm{SO}(3)$.
[For subtle reasons, it makes sense to choose the center of mass for $\boldsymbol{x}$.]

Write as deterministic plus stochastic parts:

$$\begin{pmatrix} \dot{\boldsymbol{x}} \\ \dot{\boldsymbol{\phi}} \end{pmatrix} = \begin{pmatrix} \mathbb{1} & \mathbb{0} \\ \mathbb{0} & \mathbb{L} \end{pmatrix} \cdot \left\{ \begin{pmatrix} \boldsymbol{U} \\ \boldsymbol{\Omega} \end{pmatrix} + \sqrt{2\widehat{\mathbb{D}}} \cdot \dot{\boldsymbol{w}} \right\} \qquad \text{(General ABP model)}$$

where $\dot{\boldsymbol{w}}$ is a vector of Wiener increments with correlation

$$\mathsf{E}\{\dot{\boldsymbol{w}}(t) \otimes \dot{\boldsymbol{w}}(s)\} = \delta(t-s)\, \mathbb{1} \,.$$

The fluctuation-dissipation theorem (Stokes–Einstein) implies

$$\widehat{\mathbb{D}} = k_B T \, \widehat{\mathbb{M}} \,.$$

Remark on the overdamped limit

When passing from the underdamped (Langevin) dynamics to the overdamped limit, there is a well-known stochastic drift [Lau & Lubensky (2007); Farago & Grønbech-Jensen (2014); Farago & Grønbech-Jensen (2014); Farago (2017)]:

$$\widehat{\boldsymbol{U}} = \begin{pmatrix} \boldsymbol{U} \\ \boldsymbol{\Omega} \end{pmatrix} = \begin{pmatrix} \boldsymbol{U}_{\mathrm{swim}} \\ \boldsymbol{\Omega}_{\mathrm{swim}} \end{pmatrix} + \nabla_{\widehat{\boldsymbol{x}}} \cdot \widehat{\mathbb{D}}$$

where $\widehat{\boldsymbol{x}} = (\boldsymbol{x}, \boldsymbol{\phi})$ and

$$\nabla_{\widehat{\boldsymbol{x}}} \cdot \widehat{\mathbb{D}} = \begin{pmatrix} \nabla_{\boldsymbol{x}} \cdot \mathbb{D}_{\boldsymbol{xx}} + \nabla_{\boldsymbol{\phi}} \cdot \mathbb{D}_{\boldsymbol{\phi x}} \\ \nabla_{\boldsymbol{x}} \cdot \mathbb{D}_{\boldsymbol{x\phi}} + \nabla_{\boldsymbol{\phi}} \cdot \mathbb{D}_{\boldsymbol{\phi\phi}} \end{pmatrix}$$

with $\nabla_{\boldsymbol{\phi}}$ defined appropriately for $\mathrm{SO}(3)$ $\quad (\nabla_{\boldsymbol{\phi}} := \mathbb{L}^{\top} \cdot \partial_{\boldsymbol{\phi}})$.

For a free particle in a homogeneous medium ($\widehat{\mathbb{D}} = \widehat{\mathbb{Q}} \cdot \widehat{\mathbb{D}}^{(0)} \cdot \widehat{\mathbb{Q}}^{\top}$),

$$\nabla_{\widehat{\boldsymbol{x}}} \cdot \widehat{\mathbb{D}} = \begin{pmatrix} \boldsymbol{\epsilon} : \mathbb{D}_{\boldsymbol{\phi x}} \\ 0 \end{pmatrix}, \qquad (\boldsymbol{\epsilon})_{ijk} = \epsilon_{ijk}\,,$$

which vanishes when the centers of mass and reaction coincide.

Fokker–Planck equation

Our earlier General ABP model:

$$\frac{\mathrm{d}}{\mathrm{d}t}\widehat{\boldsymbol{x}} = \begin{pmatrix} \mathbb{1} & \mathbb{0} \\ \mathbb{0} & \mathbb{L} \end{pmatrix} \cdot \left\{ \widehat{\boldsymbol{U}} + \sqrt{2\widehat{\mathbb{D}}} \cdot \dot{\boldsymbol{w}} \right\}$$

can be turned into a Fokker–Planck equation for the probability density $p(\widehat{\boldsymbol{x}}, t) = p(\boldsymbol{x}, \boldsymbol{\phi}, t)$:

$$\partial_t p = -\nabla_{\widehat{\boldsymbol{x}}} \cdot \left\{ \widehat{\boldsymbol{U}} p - \nabla_{\widehat{\boldsymbol{x}}} \cdot \left(\widehat{\mathbb{D}}\, p \right) \right\}.$$

This equation is hard to solve, being a 6-dimensional PDE.

Our next task is to get rid of the angular dependence by passing to the long time / large scale limit.

Rescaling

For a small parameter δ, effect the diffusive rescaling

$$\partial_t \to \delta^2\,\partial_t, \qquad \nabla_{\boldsymbol{x}} \to \delta\,\nabla_{\boldsymbol{x}}, \qquad \nabla_{\boldsymbol{\phi}} \to \nabla_{\boldsymbol{\phi}}.$$

(There is no such things as a "large" angle.)

As usual, we expand

$$p = p_0 + \delta\,p_1 + \delta^2\,p_2 + \cdots\,.$$

Order δ^0

The order δ^0 part of the differential operator in the FP equation is

$$\mathcal{L}p = \nabla_{\boldsymbol{\phi}} \otimes \nabla_{\boldsymbol{\phi}} : \big(\mathbb{D}_{\boldsymbol{\phi}\boldsymbol{\phi}} p\big) - \nabla_{\boldsymbol{\phi}} \cdot \big(\boldsymbol{\Omega}\, p\big).$$

At order δ^0 we must solve

$$\mathcal{L}p_0 = 0.$$

To keep things simple, assume $\mathcal{L}p_0 = 0$ only has solutions which are independent of $\boldsymbol{\phi}$ (isotropic). Then we may write

$$p_0 = \mathcal{P}(\boldsymbol{x}, t),$$

that is, our leading-order solution is some as-yet unknown function of the large-scales variables $\boldsymbol{x}$ and t.

(*i.e.*, at long times our particle randomizes its orientation completely.)

Order δ^1

At the next order in δ, we must solve

$$\mathcal{L}p_1 = \nabla_{\boldsymbol{x}} \cdot (\boldsymbol{V}\,\mathcal{P}), \qquad \boldsymbol{V} := \boldsymbol{U} - 2\nabla_{\boldsymbol{\phi}} \cdot \mathbb{D}_{\boldsymbol{\phi x}}\,.$$

By linearity, if we can solve

$$\boxed{\mathcal{L}\boldsymbol{\chi} = \boldsymbol{V}} \qquad \text{cell problem for } \boldsymbol{\chi}$$

then we can write

$$p_1 = \nabla_{\boldsymbol{x}} \cdot (\boldsymbol{V}\,\boldsymbol{\chi})$$

Let's assume for now that we've solved this.

Order δ^2

As is common in this type of problem, we don't actually need to solve for p_2. We just need to apply a solvability condition at order δ^2 to obtain a heat equation

$$\partial_t \mathcal{P} = \nabla_{\boldsymbol{x}} \otimes \nabla_{\boldsymbol{x}} : (\mathbb{D}_{\text{eff}} \mathcal{P})$$

where the effective diffusivity is

$$\mathbb{D}_{\text{eff}} = \langle \mathbb{D}_{\boldsymbol{xx}} \rangle - \text{sym} \, \langle \boldsymbol{U} \otimes \boldsymbol{\chi} \rangle$$

and angle brackets denote an average over $\mathrm{SO}(3)$.

So, in principle we can find the effective diffusivity, as long as we can solve $\mathcal{L}\boldsymbol{\chi} = \boldsymbol{V}$.

The elliptic problem

We want to solve $\mathcal{L}\boldsymbol{\chi} = \boldsymbol{V}$:

$$\mathcal{L}\boldsymbol{\chi} = \nabla_{\boldsymbol{\phi}} \otimes \nabla_{\boldsymbol{\phi}} : \big(\mathbb{D}_{\boldsymbol{\phi\phi}}\boldsymbol{\chi}\big) - \nabla_{\boldsymbol{\phi}} \cdot \big(\boldsymbol{\Omega}\,\boldsymbol{\chi}\big) = \boldsymbol{V}$$

When the particle is of axially-symmetric shape, $\mathbb{D}_{\boldsymbol{\phi\phi}} = D_{\mathrm{r}}\,\mathbb{1}$ and the second-order operator is the spherical Laplacian. (In that case ignore the ψ Euler angle.)

We can then solve the problem by expanding $\boldsymbol{V}$ in terms of spherical harmonics, which are eigenfunctions of the Laplacian.

[See for example Cates & Tailleur (2013); Sandoval (2013).]

But in general $\mathbb{D}_{\boldsymbol{\phi\phi}}$ can be essentially arbitrary.

Avoiding spherical harmonics

We can completely avoid spherical harmonics by using the magic relation

$$\mathcal{L}\mathbb{Q} = -\mathbb{Z}^{-1} \cdot \mathbb{Q}$$

where we define the positive-definite matrix

$$\begin{aligned} \mathbb{Z}^{-1} &= (\mathrm{Tr}\,\mathbb{D}_{\boldsymbol{\phi}\boldsymbol{\phi}})\,\mathbb{1} - \mathbb{D}_{\boldsymbol{\phi}\boldsymbol{\phi}} - \boldsymbol{\Omega} \cdot \boldsymbol{\epsilon} \\ &= k_B T \{(\mathrm{Tr}\,\mathbb{M}_{\boldsymbol{\phi}\boldsymbol{\phi}})\,\mathbb{1} - \mathbb{M}_{\boldsymbol{\phi}\boldsymbol{\phi}}\} - \boldsymbol{\Omega} \cdot \boldsymbol{\epsilon} \end{aligned}$$

[Obvious? It wasn't to us! Harder calculation than it looks.]

Solution to $\mathcal{L}\boldsymbol{\chi} = \boldsymbol{V}$ is thus $\boldsymbol{\chi} = -\mathbb{Z} \cdot \mathbb{Q}$.

The effective diffusivity

We omit a lot of details, but eventually we find our sought-after isotropic effective diffusivity:

$$D_{\mathrm{eff}} = \tfrac{1}{3}\,\mathrm{Tr}\,\mathbb{D}_{\boldsymbol{x}\boldsymbol{x}} + \tfrac{1}{3}\,\boldsymbol{U}\cdot\mathbb{Z}\cdot(\boldsymbol{U} - 2\nabla_{\boldsymbol{\phi}}\cdot\mathbb{D}_{\boldsymbol{\phi}\boldsymbol{x}})$$

where recall that $\boldsymbol{U}$ is the total velocity of the particle, including noise-induced drift, and

$$\mathbb{Z}^{-1} = (\mathrm{Tr}\,\mathbb{D}_{\boldsymbol{\phi}\boldsymbol{\phi}})\,\mathbb{1} - \mathbb{D}_{\boldsymbol{\phi}\boldsymbol{\phi}} - \boldsymbol{\Omega}\cdot\boldsymbol{\epsilon}\,.$$

For a hydrodynamically isotropic particle ($\mathbb{D}_{\boldsymbol{\phi}\boldsymbol{\phi}} = D_{\mathrm{r}}\,\mathbb{1}$) with $\boldsymbol{\Omega} = 0$, reduces to the 'traditional' result

$$D_{\mathrm{eff}} = \tfrac{1}{3}\,\mathrm{Tr}\,\mathbb{D}_{\boldsymbol{x}\boldsymbol{x}} + \frac{1}{6D_{\mathrm{r}}}\,\boldsymbol{U}\cdot(\boldsymbol{U} - 2\boldsymbol{\epsilon} : \mathbb{D}_{\boldsymbol{\phi}\boldsymbol{x}})$$

with potentially a correction $\boldsymbol{\epsilon} : \mathbb{D}_{\boldsymbol{\phi}\boldsymbol{x}}$.

Wobbly passive thermal particle

There is an insight here for the most classical of all particles, a thermal passive particle ($\boldsymbol{U}_{\rm swim} = \boldsymbol{\Omega}_{\rm swim} = 0$), which still has a noise-induced drift:

$$\boldsymbol{U} = \nabla_{\boldsymbol{\phi}} \cdot \mathbb{D}_{\boldsymbol{\phi x}} = \boldsymbol{\epsilon} : \mathbb{D}_{\boldsymbol{\phi x}}\,, \qquad \boldsymbol{\Omega} = \nabla_{\boldsymbol{\phi}} \cdot \mathbb{D}_{\boldsymbol{\phi\phi}} = 0.$$

The term $\boldsymbol{\epsilon} : \mathbb{D}_{\boldsymbol{\phi x}}$ can only be nonzero if the centers of mass and reaction don't coincide.

When subjected to noise, such a 'wobbly passive particle' behaves a bit like an ABP, with an effective noise-induced swimming velocity!

Does this have consequences?

Passive thermal particle: Effective diffusivity

Part of the original motivation for this work was a claim in a recent preprint that the noise-induced drift $\nabla_{\boldsymbol{\phi}} \cdot \mathbb{D}_{\boldsymbol{\phi x}}$ for a wobbly particle leads to an 'enhanced' effective diffusivity

$$D_{\text{eff}} = \tfrac{1}{3}\,\mathrm{Tr}\,\mathbb{D}^{\mathrm{h}}_{\boldsymbol{xx}} \times \left(1 + \tfrac{1}{2}\,\|\Delta\boldsymbol{x}\|^2/a^2\right) \qquad \text{wobbliness correction}$$

where $\mathbb{D}^{\mathrm{h}}_{\boldsymbol{xx}} = k_B T\,\mathbb{M}^{\mathrm{h}}_{\boldsymbol{xx}}$ is the diffusivity tensor defined from the center of hydrodynamic reaction, *i.e.*, as if the particle was not wobbly.

However, accounting for the coupling terms between the rotational and translational degrees of freedom reveals

$$\boxed{D_{\text{eff}} = \tfrac{1}{3}\,\mathrm{Tr}\,\mathbb{D}^{\mathrm{h}}_{\boldsymbol{xx}}}$$

Conclude: even though the small-scale dynamics differ, the large-scale dynamics of a wobbly particles are the same as an unwobbly one.

Other results and future work

- Previous work called 'chiral' a particle with a net rotation $\mathbf{\Omega}$, but the particle was axially symmetric.
- Here we can involve chirality through shape as well.
- Can derive a more complex, general formula for particles subjected to external field (Sevilla, 2016).
- Can also allow dependence large-scale variables that can lead to a novel large-scale drift.
- Our work actually focuses also on non-thermal active particles, involving a generalized fluctuation-dissipation theorem.
- We derive the overdamped limit from the Langevin equation, which is involves subtleties regarding the interpretation of multiplicative noise.
- 2D version is published: Thiffeault, J.-L. & Guo, J. (2022). *Phys. Rev. E,* **106** (1), L012603

References I

Ai, B., Chen, Q., He, Y., Li, F., & Zheng, W. (2013). *Phys. Rev. E,* **88**, 062129.

Baskaran, A. & Marchetti, M. (2008). *Phys. Rev. E,* **77**, 011920.

Cates, M. E. & Tailleur, J. (2013). *Europhys. Lett.* **101**, 20010.

Chen, H. & Thiffeault, J.-L. (2021). *J. Fluid Mech.* **916**, A15.

Farago, O. (2017). *Phys. Rev. E,* **96** (4).

Farago, O. & Grønbech-Jensen, N. (2014). *J. Stat. Phys.* **156**, 1093–1110.

Farago, O. & Grønbech-Jensen, N. (2014). *Phys. Rev. E,* **89** (1).

Happel, J. & Brenner, H. (1983). *Low Reynolds number hydrodynamics*. The Hague, Netherlands: Martinus Nijhoff (Kluwer).

Kurzthaler, C. & Franosch, T. (2017). *Soft Matter,* **13**, 6396–6406.

Kurzthaler, C., Leitmann, S., & Franosch, T. (2016). *Sci. Rep.* **6**, 36702.

Lau, A. W. C. & Lubensky, T. C. (2007). *Phys. Rev. E,* **76**, 0111123.

Peruani, F. & Morelli, L. G. (2007). *Phys. Rev. Lett.* **99** (1).

Redner, G. S., Hagan, M. F., & Baskaran, A. (2013). *Phys. Rev. Lett.* **110**, 055701.

Romanczuk, P., Bär, M., Ebeling, W., Lindner, B., & Schimansky-Geier, L. (2012). *The European Physical Journal Special Topics,* **202** (1), 1–162.

Romanczuk, P. & Schimansky-Geier, L. (2011). *Phys. Rev. Lett.* **106** (23).

References II

Sandoval, M. (2013). *Phys. Rev. E,* **87** (3).

Sevilla, F. J. (2016). *Phys. Rev. E,* **94** (6).

Solon, A. P., Fily, Y., Baskaran, A., Cates, M. E., Kafri, Y., Kardar, M., & Tailleur, J. (2015). *Nat. Phys.* **11**, 673–678.

Stenhammar, J., Marenduzzo, D., Allen, R., & Cates, M. E. (2014). *Soft Matter,* **10**, 1489–1499.

Thiffeault, J.-L. & Guo, J. (2022). *Phys. Rev. E,* **106** (1), L012603.

van Teeffelen, S. & Löwen, H. (2008). *Phys. Rev. E,* **78**, 020101.

Wagner, C. G., Hagan, M. F., & Baskaran, A. (2017). *J. Stat. Mech.: Theory Exp.* **2017** (4), 043203.

Zöttl, A. & Stark, H. (2016). *J. Phys.: Condens. Matter,* **28**, 253001.