Supporting Information

Internal gradient distributions: A susceptibility-derived tensor delivering morphologies by magnetic resonance

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Supporting Information 1: A formal derivation of the internal gradient distribution tensor

The normalized magnetization arising from an ensemble of non-interacting and equivalent spins under the effects of a sequence of pulses or modulating gradients is $M(t) = \left\langle e^{-i\phi(t)}\right\rangle$, where the brackets account for an ensemble average over the random phases $\phi(t)$. For the spin-echo sequences being considered in this work, the average phase $\langle\phi(t)\rangle$ will be equal to zero. Assuming that the random phase $\phi(t)$ has a Gaussian distribution [48], $M(t) = \exp\left\{-\frac{1}{2}\langle\phi^2(t)\rangle\right\}$, the signal will evidence a decay depending on the attenuation factor $\beta(t) = \frac{1}{2}\langle\phi^2(t)\rangle$. With most sources of decoherence normalized out by the constant-time, constant-pulses-number, fixed-number-of-gradients nature of the NOGSE sequences assayed [20, 21, 44], we ascribe to diffusion effects as the sole source of this attenuation. It is then convenient to describe the β-factor in terms of the gradient modulating function $\vec{G}_{\rm tot}(t')$ [45–47]:

$$\beta(TE) = \frac{\gamma^2}{2}\int_0^{TE} dt' \int_0^{TE} dt'' \vec{G}^{\dagger}_{\rm tot}(t')\cdot\langle\vec{r}(t')\vec{r}(t'')\rangle\cdot\vec{G}_{\rm tot}(t'') \quad \text{(S.1)}$$

$$= \frac{\gamma^2}{2}\int_{-\infty}^{\infty} dt' \int_{-\infty}^{\infty} dt'' \vec{G}^{\dagger}_{\rm tot}(t',TE)\cdot\mathbf{g}\left(t''-t'\right)\cdot\vec{G}_{\rm tot}(t'',TE), \quad \text{(S.2)}$$

where in the second equation we redefined the gradient modulation function such that $\vec{G}_{\rm tot}(t',TE) = 0$ if $t' < 0$ or $t' > TE$ (i.e., outside the total evolution time range). The evolution is given in terms of a tensorial correlation function reflecting the displacements' fluctuations $\mathbf{g}(\tau) = \langle\Delta\vec{r}(t')\Delta\vec{r}(t'+\tau)\rangle$; i.e. $g_{i,j} = \langle\Delta x_i(t')\Delta x_j(t'+\tau)\rangle$ with i,j representing the spatial axis x, y, z. This correlation function can be related to a diffusion power spectrum $\mathbf{D}(\omega)$ [4, 5, 45, 46] by a Fourier transform: $\mathcal{FT}\{\mathbf{g}(\tau)\}/\sqrt{2\pi} = \mathbf{D}(\omega)/\omega^2$. In the event of anisotropic diffusion, Eq. (S.1) can thus be recast in its Fourier representation [45–47] as:

$$\beta(TE) = \frac{\gamma^2}{2}\int_{-\infty}^{\infty} d\omega \vec{\mathcal{G}}^{\dagger}_{\rm tot}(\omega,TE)\cdot\frac{\mathbf{D}(\omega)}{\omega^2}\cdot\vec{\mathcal{G}}_{\rm tot}(\omega,TE), \quad \text{(S.3)}$$

where $\vec{\mathcal{G}}_{\rm tot}(\omega, TE) = \vec{\mathcal{G}}(\omega, TE) + \vec{\mathcal{G}}_0\,(\omega, TE)$, is the filter function introduced in Eq. (1) of the main text.

Considering the applied gradient modulation $\vec{G}(t', TE)$, the internal background gradient modulation $\vec{G}_0(t', TE)$, and their respective filter functions $\vec{\mathcal{G}}(\omega, TE)$ and $\vec{\mathcal{G}}_0\,(\omega, TE)$, the argument of the integral defining this attenuation factor can then be expanded as

$$\begin{aligned}\vec{\mathcal{G}}^{\dagger}_{\rm tot}(\omega, TE)\cdot\frac{\mathbf{D}(\omega)}{\omega^2}\cdot\vec{\mathcal{G}}_{\rm tot}(\omega, TE) = &\underbrace{\vec{\mathcal{G}}^{\dagger}(\omega, TE)\cdot\frac{\mathbf{D}(\omega)}{\omega^2}\cdot\vec{\mathcal{G}}(\omega, TE)}_{\text{external gradient dephasing}}\\ &+\underbrace{\vec{\mathcal{G}}^{\dagger}_0(\omega, TE)\cdot\frac{\mathbf{D}(\omega)}{\omega^2}\cdot\vec{\mathcal{G}}_0(\omega, TE)}_{\text{internal gradient dephasing}}\\ &+\underbrace{2\Re\left\{\vec{\mathcal{G}}^{\dagger}(\omega, TE)\cdot\frac{\mathbf{D}(\omega)}{\omega^2}\cdot\vec{\mathcal{G}}_0\,(\omega, TE)\right\}}_{\text{cross-term}}.\end{aligned}\qquad\text{(S.4)}$$

This leads to Eq. (2) of the main text, where $\beta(TE) = \beta_{G^2}(TE) + \beta_{G_0^2}(TE) + \beta_{\vec{G}\cdot\vec{G}_0}(TE)$ and the normalized spin magnetization becomes

$$M(TE) = M_{G^2}(TE) \times M_{G_0^2}\,(TE) \times M_{\vec{G}.\vec{G}_0}(TE). \qquad\text{(S.5)}$$

Assuming a $\vec{G}(t', TE) = \vec{G}\,f(t', TE)$, involving a strength vector $\vec{G}$ and a time-dependency $f(t', TE)$, then $\vec{\mathcal{G}}(\omega, TE) = \vec{G}\,F(\omega, TE)$ with $F(\omega, TE)$ the Fourier transform of $f(t', TE)$. The applied gradient diffusion attenuation becomes

$$M_{G^2}(TE) = \exp\left\{-\beta_{G^2}(TE)\right\}, \qquad\text{(S.6)}$$

where

$$\beta_{G^2}(TE) = \tfrac{\gamma^2 G^2}{2}\int_{-\infty}^{\infty} d\omega\,\frac{D_G(\omega)}{\omega^2}\,|F(\omega, TE)|^2\,, \qquad\text{(S.7)}$$

and $D_G(\omega) = \left[\vec{G}^{\dagger}\cdot\mathbf{D}(\omega)\cdot\vec{G}\right]/G^2$. Likewise, the pure background gradient decay is independent of the applied gradient

$$M_{G_0^2}\,(TE) = \exp\left\{-\beta_{G_0^2}\,(TE)\right\}, \qquad\text{(S.8)}$$

where

$$\beta_{G_0^2}(TE, \vec{G}_0) = \tfrac{\gamma^2 G_0^2}{2} \int_{-\infty}^{\infty} d\omega \frac{D_{G_0}(\omega)}{\omega^2} \left| F_0(\omega, TE) \right|^2, \tag{S.9}$$

with $D_{G_0}(\omega) = \left[\vec{G}_0^\dagger \cdot \mathbf{D}(\omega) \cdot \vec{G}_0 \right] / G_0^2$, and we have again assumed that $\vec{G}_0(t', TE) = \vec{G}_0 \, f_0(t', TE)$ and thereby $\vec{\mathcal{G}}_0(\omega, TE) = \vec{G}_0 \, F_0(\omega, TE)$. Finally, the cross-term attenuation will be

$$\beta_{\vec{G}\cdot\vec{G}_0} \left(TE, \vec{G}, \vec{G}_0 \right) = \tfrac{\gamma^2}{2} \vec{G}^\dagger \cdot \left[\int_{-\infty}^{\infty} d\omega \, 2\mathrm{Re} \left\{ F^\dagger(\omega, TE) \, \frac{\mathbf{D}(\omega)}{\omega^2} \, F_0 \left(\omega, TE \right) \right\} \right] \cdot \vec{G}_0 = \vec{G}^\dagger \cdot \tilde{\mathbf{D}} \cdot \vec{G}_0, \tag{S.10}$$

where $\tilde{\mathbf{D}} = \tfrac{\gamma^2}{2} \left[\int_{-\infty}^{\infty} d\omega \, 2\mathrm{Re} \left\{ F^\dagger(\omega, TE) \, \frac{\mathbf{D}(\omega)}{\omega^2} \, F_0 \left(\omega, TE \right) \right\} \right]$.

Our derivations also assumed that $\vec{G}_0$ can be described by a Gaussian distribution. The cross-term contribution to the attenuation factor turns out to be

$$\beta_{\vec{G}\cdot\vec{G}_0} \left(TE \right) = \vec{G}^\dagger \cdot \tilde{\mathbf{D}} \cdot \left\langle \vec{G}_0 \right\rangle + \vec{G}^\dagger \cdot \tilde{\mathbf{D}} \cdot \left\langle \Delta\vec{G}_0 \Delta\vec{G}_0 \right\rangle \cdot \tilde{\mathbf{D}} \cdot \vec{G}, \tag{S.11}$$

where $\Delta\vec{G}_0 = \vec{G}_0 - \left\langle \vec{G}_0 \right\rangle$ and $\left\langle \Delta\vec{G}_0 \Delta\vec{G}_0 \right\rangle$ is the internal gradient-distribution tensor (IGDT). This second term is always positive, since it is a quadratic term, while the first term depends on the relative sign of the parallel component of $\left[\vec{G}^\dagger \cdot \tilde{\mathbf{D}} \right]_{\parallel}$ to the background gradient G_0.

For an isotropic diffusion $\mathbf{D}(\omega) = D(\omega)\mathbf{I}$, the attenuation factor get the simplified form

$$\beta_{\vec{G}\cdot\vec{G}_0} \left(TE \right) = \tilde{D}_{\mathrm{iso}} \, \vec{G}^\dagger \cdot \left\langle \vec{G}_0 \right\rangle + \tilde{D}_{\mathrm{iso}}^2 \, \vec{G}^\dagger \cdot \left\langle \Delta\vec{G}_0 \Delta\vec{G}_0 \right\rangle \cdot \vec{G}, \tag{S.12}$$

where $\tilde{D}_{\mathrm{iso}} = \tfrac{\gamma^2}{2} \left[\int_{-\infty}^{\infty} d\omega \, 2\mathrm{Re} \left\{ F^\dagger(\omega, TE) \, \frac{D(\omega)}{\omega^2} \, F_0 \left(\omega, TE \right) \right\} \right]$.

As an example on the use of this formalism, we consider the sequence of Fig. 1 of the main text and assume free diffusion to derive Eq. (3-5) of the main text. For free diffusion only the tail of the displacement power spectrum $D(\omega) \propto 1/\omega^2$ is important [20, 44]. The purely applied-gradient diffusion term $M_{G^2}(TE)$ is as derived for a CPMG sequence [44]

$$M_{G^2}(TE) = \exp\left\{ -\frac{1}{12} \gamma^2 G^2 D_0 \frac{TE^3}{N^2} \right\}, \tag{S.13}$$

where the delay $x = TE/N$. The pure background gradient decay term is in turn the one that corresponds to a spin-echo modulation [44]

$$M_{G_0^2}\left(TE, N \right) = \exp\left\{ -\frac{1}{12} \gamma^2 G_0^2 D_0 TE^3 \right\}, \tag{S.14}$$

which is independent of x. The cross-term signal-decay contribution is calculated from Eq. (S.10) leading to

$$M_{\vec{G}\cdot\vec{G}_0}\left(TE\right) = \exp\left\{\frac{1}{4}\gamma^2\vec{G}\cdot\vec{G}_0 D_0 \frac{TE^3}{N^2}\right\}. \tag{S.15}$$

Supporting Information 2: sNOGSE/aNOGSE's: Analytical attenuation expressions for the general case of anisotropic diffusion

We calculate next the normalized spin signal arising from Eq. (S.5),

$$M^{\binom{s}{a}NOGSE}(TE) = M_{G^2}^{\binom{s}{a}NOGSE}(TE) \times M_{G_0^2}\left(TE\right) \times M_{\vec{G}.\vec{G}_0}^{\binom{s}{a}NOGSE}(TE), \tag{S.16}$$

for the symmetric and asymmetric non-uniform gradient spin echo modulations ($\binom{\mathrm{s}}{\mathrm{a}}$NOGSE) introduced in Fig. 2. As described in the main text,

$$M_{G^2}^{sNOGSE}(TE) = M_{G^2}^{aNOGSE}(TE) \tag{S.17}$$

as a result of

$$F^{\mathrm{sNOGSE}}(\omega, TE) = F^{\mathrm{aNOGSE}}(\omega, TE) \tag{S.18}$$

in Eq. (S.7). The pure background gradient signal contribution is therefore independent of the applied gradient modulation and direction, providing the same weight for both NOGSE sequences. The cross-term in the attenuation factor for sNOGSE is zero but that for aNOGSE is not, as the products $F^{sNOGSE\dagger}(\omega, TE)\, F_0\left(\omega, TE\right)$ and $F^{aNOGSE\dagger}(\omega, TE)\, F_0\left(\omega, TE\right)$ in Eq. (S.10) are odd and even functions of ω, respectively. This cross-term between the aNOGSE-modulated applied gradient and the background gradient G_0 will be

$$\beta_{\vec{G}\cdot\vec{G}_0}\left(TE\right) = \tfrac{\gamma^2}{2}\vec{G}\cdot\left[\int_{-\infty}^{\infty} d\omega\, 2\mathrm{Re}\left\{\left(F^{aNOGSE}(\omega, TE)\right)^{\dagger} \frac{\mathbf{D}(\omega)}{\omega^2} F_0\left(\omega, TE\right)\right\}\right]\cdot\vec{G}_0 = \vec{G}\cdot\tilde{\mathbf{D}}\cdot\vec{G}_0. \tag{S.19}$$

As explained in the main text, the measured spin signal decays for the sNOGSE and aNOGSE sequences as described in Fig. 2e, factor out all non-diffusing sources of decoherence after normalizing them by the single-echo signal [20, 21, 44]. The amplitude of the

NOGSE modulation is then

$$M_{CPMG}(TE)/M_{Single-echo}(TE) = \exp\left(-\Delta\beta\right) = \exp\left[-\left(\beta^{CPMG} - \beta^{Single-echo}\right)\right], \quad \text{(S.20)}$$

where the amplitude contrast of the attenuation factors $\Delta\beta = \beta^{CPMG} - \beta^{Single-echo}$. As the contribution to the attenuation factor that purely depends of the background gradient is independent of the applied gradient modulation its contribution $\Delta\beta_{G_0^2}$ is null, and the amplitude of the attenuation factors is then

$$\Delta\beta = \Delta\beta_{G^2} + \Delta\beta_{\vec{G}\cdot\vec{G}_0}. \quad \text{(S.21)}$$

For the sNOGSE sequence $\Delta\beta^s = \Delta\beta_{G^2}$ as the cross-term is null, and $\Delta\beta^a = \Delta\beta_{G^2} + \Delta\beta_{\vec{G}\cdot\vec{G}_0}$ for the aNOGSE modulation curve. Notice that the contribution of the term that only depends of the applied gradient $\Delta\beta_{G^2}$ is the same for both sequences according to Eqs. (S.17) and (S.18). Then by subtracting $\Delta\beta^a$ and $\Delta\beta^s$, the $\Delta\beta_{\vec{G}\cdot\vec{G}_0}$ cross-term contribution to the amplitude modulation is obtained, where

$$\Delta\beta_{\vec{G}\cdot\vec{G}_0} = \frac{\gamma^2}{2}\vec{G}\cdot\left[\int_{-\infty}^{\infty} d\omega\, 2\mathrm{Re}\left\{\left[F^{aNOGSE}(\omega,TE) - F^{sNOGSE}(\omega,TE)\right]^\dagger \frac{\mathbf{D}(\omega)}{\omega^2} F_0\left(\omega,TE\right)\right\}\right]\cdot\vec{G}_0 \quad \text{(S.22)}$$

$$= \vec{G}\cdot\Delta\tilde{\mathbf{D}}\cdot\vec{G}_0, \quad \text{(S.23)}$$

with $\Delta\tilde{\mathbf{D}} = \frac{\gamma^2}{2}\int_{-\infty}^{\infty} d\omega\, 2\mathrm{Re}\left\{\left[F^{aNOGSE}(\omega,TE) - F^{sNOGSE}(\omega,TE)\right]^\dagger \frac{\mathbf{D}(\omega)}{\omega^2} F_0\left(\omega,TE\right)\right\}$. Assuming as before a Gaussian distribution for G_0,

$$\Delta\beta_{\vec{G}\cdot\vec{G}_0} = \vec{G}\cdot\Delta\tilde{\mathbf{D}}\cdot\left\langle\vec{G}_0\right\rangle + \vec{G}\cdot\Delta\tilde{\mathbf{D}}\cdot\left\langle\Delta\vec{G}_0\Delta\vec{G}_0\right\rangle\cdot\Delta\tilde{\mathbf{D}}\cdot\vec{G}. \quad \text{(S.24)}$$

where the first term depends on the relative sign of the parallel component of $\left[\vec{G}\cdot\mathbf{D}(\omega)\right]_{\parallel}$ to the background gradient G_0, which depends of the anisotropic restricted-diffusion weighting. Notice that the second term is always positive and it contains the IGDT $\left\langle\Delta\vec{G}_0\Delta\vec{G}_0\right\rangle$. This was the expression used to evaluate the results presented in Fig. 4 after being normalized by $\Delta\beta^s = \Delta\beta_{G^2}$ to remove the anisotropic weighting due to restricted diffusion effects. For an isotropic diffusion $\mathbf{D}(\omega) = D(\omega)\mathbf{I}$, this gets simplified to

$$\Delta\beta_{\vec{G}\cdot\vec{G}_0} = \Delta\tilde{D}_{\mathrm{iso}}\vec{G}\cdot\left\langle\vec{G}_0\right\rangle + \Delta\tilde{D}_{\mathrm{iso}}^2\vec{G}\cdot\left\langle\Delta\vec{G}_0\Delta\vec{G}_0\right\rangle\cdot\vec{G}. \quad \text{(S.25)}$$

where $\Delta\tilde{D}_{\mathrm{iso}} = \frac{\gamma^2}{2}\int_{-\infty}^{\infty} d\omega\, 2\mathrm{Re}\left\{\left[F^{aNOGSE}(\omega,TE) - F^{sNOGSE}(\omega,TE)\right]^\dagger D(\omega)\, F_0\left(\omega,TE\right)\right\}$.

Supporting Information 3: sNOGSE/aNOGSE analytical expressions for the case of free, unrestricted diffusion

For free diffusion only the tail of the displacement power spectrum $D(\omega) \propto 1/\omega^2$ will be important, as it is this short-times regime active before restriction effects are seen, that matters [20, 44]. The purely applied-gradient diffusion term $M_{G^2}^{\binom{s}{a}NOGSE}(TE)$ is then given by [44]

$$M_{G^2}^{\binom{\mathrm{s}}{\mathrm{a}}\mathrm{NOGSE}}(TE) = \exp\left\{-\frac{1}{12}\gamma^2 G^2 D_0 \left[(N-2)\,x^3 + 2y^3\right]\right\}, \tag{S.26}$$

where $(N-2)\,x + 2y = TE_{NOGSE} = TE/2$ (see Fig. 2 of the main text for definitions). The pure background gradient decay term is in turn

$$M_{G_0^2}\left(TE, N\right) = \exp\left\{-\frac{1}{12}\gamma^2 G_0^2 D_0 TE^3\right\}, \tag{S.27}$$

which is independent of x and y, and therefore of the applied gradient modulation as was mentioned in the manuscript.

The cross-term signal-decay contribution for sNOGSE is zero as described before, and the one for aNOGSE will be

$$M_{\vec{G}\cdot\vec{G}_0}^{aNOGSE}\left(TE\right) = \exp\left\{-\frac{1}{4}\gamma^2 \vec{G}\cdot\vec{G}_0 D_0 TE_{NOGSE}\left(-1\right)^{N/2}\left[2y^2 - \left(1 + (-1)^{N/2}\right)x^2\right]\right\}. \tag{S.28}$$

Notice that the sign of the attenuation factor for this cross-term contribution depends of the relative sign of $G_{\parallel} G_0$, where $G_{\parallel}$ is the applied component of the G-gradient that is parallel to the background gradient vector. The extremes of this attenuation arise for $x = y = TE_{NOGSE}/N$ (CPMG-like modulation)

$$\beta_{\vec{G}\cdot\vec{G}_0}^{CPMG}\left(TE\right) = -\frac{1}{4}\gamma^2 \vec{G}\cdot\vec{G}_0 D_0 \frac{TE_{NOGSE}^3}{N^2}\left((-1)^{N/2} - 1\right), \tag{S.29}$$

and for $y = TE_{NOGSE}/2$ and $x = 0$ (single-echo modulation)

$$\beta_{\vec{G}\cdot\vec{G}_0}^{Single-echo}\left(TE\right) = -\frac{1}{8}\gamma^2 \vec{G}\cdot\vec{G}_0 D_0 TE_{NOGSE}^3\left(-1\right)^{N/2}. \tag{S.30}$$

Given the $(-1)^{N/2}$ factor in Eq. (S.28) it follows that if $N/2$ is even, $\beta_{\vec{G}\cdot\vec{G}_0}^{CPMG}(TE) = 0$ and the contrast contribution for the difference of attenuation factors is

$$\Delta\beta_{\vec{G}\cdot\vec{G}_0} = \frac{1}{8}\gamma^2 \vec{G}\cdot\vec{G}_0 D_0 TE_{NOGSE}^3; \tag{S.31}$$

i.e., it depends on the relative sign of $G_{\parallel}$ and G_0. If $N/2$ is odd $\beta^{CPMG}_{\vec{G}\cdot\vec{G}_0}(TE) \neq 0$; but for $N/2$ odd the attenuation decays with $1/N^2$, and this makes the contrast lower. Assuming a Gaussian distribution for G_0,

$$\begin{aligned}\beta_{\vec{G}\cdot\vec{G}_0}(TE) = &-\frac{1}{4}\gamma^2 D_0 TE_{NOGSE}\,(-1)^{N/2}\,(2y^2 - \left(1+(-1)^{N/2}\right)x^2)\,\vec{G}\cdot\left\langle\vec{G}_0\right\rangle \\ &+\frac{1}{32}\gamma^4 D_0^2 TE_{NOGSE}^2\left[2y^2-\left(1+(-1)^{N/2}\right)x^2\right]^2\left\langle\left[\vec{G}\cdot\left(\vec{G}_0-\left\langle\vec{G}_0\right\rangle\right)\right]^2\right\rangle. \quad \text{(S.32)}\end{aligned}$$

The attenuation factor contrast amplitude is then

$$\Delta\beta_{\vec{G}\cdot\vec{G}_0} = \frac{1}{8}\gamma^2 D_0 TE^3_{NOGSE}\,\vec{G}\cdot\left\langle\vec{G}_0\right\rangle + \frac{1}{128}\gamma^4 D_0^2 TE^6_{NOGSE}\,\vec{G}\cdot\left\langle\Delta\vec{G}_0\Delta\vec{G}_0\right\rangle\cdot\vec{G}, \quad \text{(S.33)}$$

where $\left\langle\Delta\vec{G}_0\Delta\vec{G}_0\right\rangle$ is the IGDT.

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