Autocorrelation Spectra of Air-Fluidized Granular System by the NMR Spin Echo

Janez Stepišnik & Samo Lasič & Aleš Mohorič & Igor Serša & Gorazd Planinšič
University of Ljubljana, Faculty of Mathematics and Physics, Physics Department and Institute Josef Stefan, Ljubljana, Slovenia

NMR technique of modulated gradient spin echo makes possible to measure the power spectra of displacement and velocity autocorrelation functions of the air-fluidized granular beds. The obtained frequency distribution of displacement and velocity spectra at different degrees of fluidization leads to an empiric formula that well-fits to the experimental data and gives negative long-range tail of velocity autocorrelation function with a clear indication of grain caging by its neighbors and grain hopping between cagings.

1 INTRODUCTION

Most theoretical efforts of granular dynamics start by considering the fluidized granular medium as a dense, inelastic gas with a temperature defined by local velocity fluctuations (Bagnold 1954; Jenkins and Savage 1983). However, there are two particularly important aspects that contribute to the unique properties of granular materials: ordinary temperature plays no role, and the interactions between grains are dissipative because of static friction and the inelasticity of collisions. In order to reveal the underlying nature of the dynamical processes, the velocity autocorrelation function (VAF)is a key quantity from which basic transport properties can be calculated. Its knowledge gives the understanding of macroscopic properties in terms of single grain dynamics.

The experimental techniques used to study these systems span a wide range of approaches and sophistication. From the examination of spots left by carbon paper to high speed videography, magnetic resonance methods, x-ray tomography, diffusing-wave spectroscopy, positron emission method (PE) etc. According to our best knowledge only PE particle tracking was able to glimpse directly into VAF of highly fluidized granular beds (Wildman et al. 2002) that exhibits a non-Eskog decay.

2 CONCEPTUAL MODEL

We report about the first measurement of autocorrelation spectra of grain motion in the fluidized system by the NMR method of modulated gradient spin echo (MGSE) With the MGSE methods, the repetitive train of RF pulses with interspersed magnetic field gradient pulses or waveform periodically modulates the spin phase and gives the spin-echo attenuation proportional to a value of power spectra of displacement (DPS) or of power spectra of velocity autocorrelation function (VPS), depending on pulse sequence (Stepišnik 1981; Callaghan and Stepišnik 1996). The rate of the spin phase modulation determines the frequency range of measurement, which is between a few Hz to about 100kHz at present state of art, which is about the range of beads motion in fluidized granular systems according to Refs.(Menon and Durian 1997a; Menon and Durian 1997b). MGSE method can be considered as a low frequency complement of the non-elastic neutron scattering that covers the frequency range above 1 GHz.

The spin echo attenuation of spin-bearing particles moving in a weak magnetic field gradient can be written as the integral of two overlapping spectra (Stepišnik 1981; Stepišnik 1985),

$$\beta(\tau) = \frac{\gamma^2}{\pi} \int_0^\infty I_z(\omega) |G(\omega,\tau)|^2 \, d\omega. \qquad (1)$$

$I_z(\omega)$ is the power spectrum , which is according to Wiener-Khintchine teorem (Kubo 1959) related to displacement autocorrelation function along applied gradient,$\Delta z = z - \langle z \rangle$,as

$$I_z(\omega) = \int_0^\infty \langle \Delta z(t)\, \Delta z(0) \rangle e^{-i\omega t} dt, \qquad (2)$$

While, the gradient spectrum

$$\mathbf{G}(\omega,\tau) = \int_0^\tau \mathbf{G}_{eff}(t) e^{-i\omega t} \, dt, \qquad (3)$$

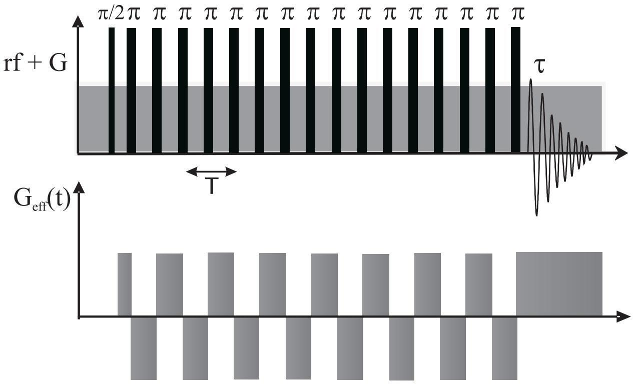

Figure 1: The sequence of π radiofrequency pulses applied to the spins in the constant magnetic field creates a tilting effective gradient.

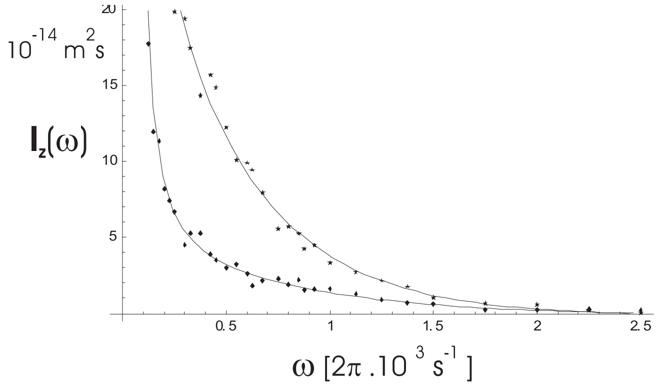

Figure 2: Displacement auto-correlation spectra of the oil-filled beds fluidized by air blowing at different pressure (p=0.25 bar) (small dots) and p=0.5 bar).The experimental points exhibits clear exponential decay as frequency increases, but follows close to $\frac{1}{\omega^2}$-dependence at small frequencies. Eq.6 gives a good fit in whole range of observation (curve).

is the Fourier transform of the effective magnetic gradient as appears to the spins, which are subjected to the combined sequence of gradient and radiofrequency pulses.

The train of π RF pulses applied to spins in the constant magnetic field gradient, as shown in Fig.1, brings about the spectrum of effective gradient that has the dominant sampling peak at the frequency $\omega = \pm\omega_m = \pm 2\pi/T$. It gives the spin echo attenuation

$$\beta(NT) \;=\; NT\frac{8\gamma^2\mathbf{G}^2}{\pi^2}I_z(\omega_m), \qquad (4)$$

which can be used to sample $I_z(\omega_m)$ by varying the interval T (Stepišnik and Callaghan 2000; Callaghan and Stepišnik 1996; Callaghan and Stepišnik 1995). By knowing $I_z(\omega_m)$, we can obtain the mean squared displacement of spin bearing particle from the relation

$$\langle \Delta z(t)^2 \rangle = \frac{4}{\pi}\int_0^\infty I_z(\omega)(1-\cos(\omega t))d\omega \qquad (5)$$

as well as the power spectra of particle velocity fluctuation using the relation $D(\omega) = I_z(\omega)\omega^2$ (Kubo 1959).

3 MEASUREMENT & ANALYSIS

The granular system of pharmaceutical 3-mm oil-filled, hard plastic spherical beads in the cylindrical container was fluidized by injecting the air through the holes in the walls. Degree of fluidization was regulated by the air pressure. The experiment was carried out using a TecMag NMR spectrometer with a horizontal bore $2.35T$ superconductive magnet equipped with micro-imaging accessories. Fig.2 shows $I_z(\omega)$ of air-fluidized beds as follows from the spin echo attenuation measurements according to Eq.4. For different gas pressure, the spectra exhibit an exponential decline for the frequencies above 400Hz, what is particularly distinctive for fluidization at high gas pressure of 0.5 bar. However, the fast grain motion amplify higher frequencies but limits the examination below 300Hz due to the strong spin echo attenuation. In order to trace $I_z(\omega)$ below 400Hz, the air blow is reduced to 0.25 bar. In this low frequency range, the exponential distribution of $I_z(\omega)$ passes into a kind of $1/\omega^2$-dependence. The power spectrum of velocity fluctuation, $D(\omega)$ follows from $I_z(\omega)$ by using well-known interrelation ,Fig.3 shows VPS, which is not of the Lorentzian-type as expected from the Enskog exponential decay of VAF, but exhibits a lobe more like the over-damped thermal harmonic oscillators (Wang and Ornstein 1945). Interestingly, the positron emission measurement of the VAF of vibro-fluidized granular beds (Wildman et al. 2002) gave very similar lobe, which was explained by the constraining nature of the experimental cell.

Good signal to noise ratio of the MGSE measurements, particularly in the low frequencies range, permits detailed analysis of $I_z(\omega)$ and $D(\omega)$ of the fluidized granular system. Fig.3B shows a clear ω^2- dependence of VPS at low frequencies, which is typical for the molecular diffusion in constrained media, where the intersection of $D(\omega)$ with the ordinate gives diffusion hopping between confinements. The model of random and ballistic grain motion between successive collisions, in which many collisions are required for a grain to break out of its cage of nearest neighbors, seems to be similar to the model of molecular restricted diffusion, where the inter-pore channels permits the motion across the system. Along this line, we can define the characteristic grain displacement within the cage, $\langle\xi^2\rangle$, the mean collisions time τ_c and the diffusion like constant D that denotes the grain motion between different cagings. By replacing the Lorentzian frequency distribution $I_z(\omega)$ of the restricted diffusion case (Wang and Ornstein 1945) by the exponential one as sugested by our measurements, we come to an empirical formula for $I_z(\omega)$ of flu-

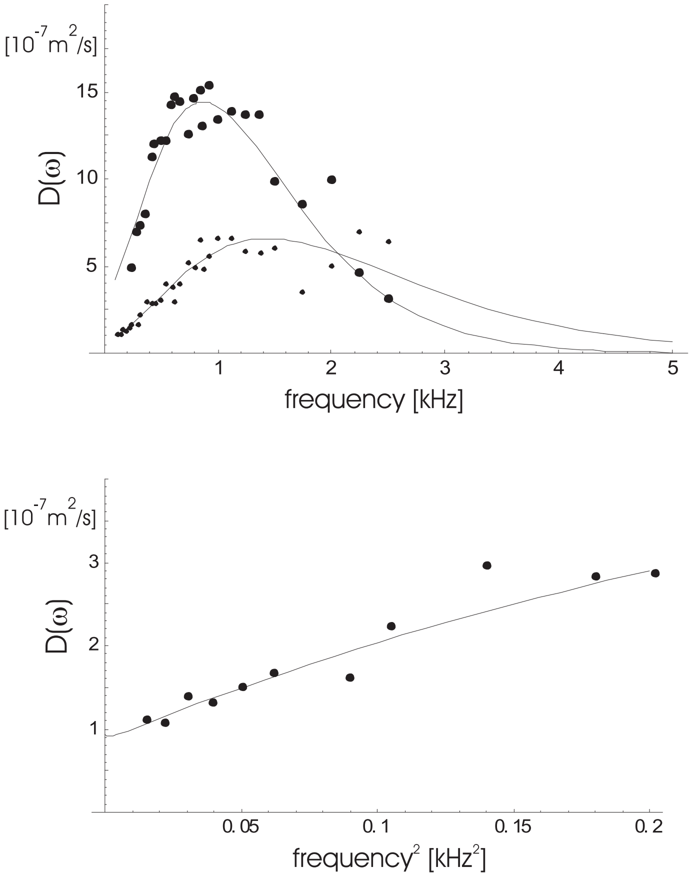

Figure 3: Power spectra of velocity autocorrelation function of the oil-filled beds fluidized by air blow at different pressures (0.25 bar and 0.5 bar). In the ω^2 dependence, the intersection of fitted curve with the ordinate gives the diffusion-like constant of grains.

idized granular system as

$$I_z(\omega) = \frac{D - \langle \xi^2 \rangle \tau_c \omega^2}{\omega^2} e^{-\tau_c \omega} \tag{6}$$

The calculated mean squared displacement according to Eq,5 gives the time dependence as reported from other measurements and simulations (Menon and Durian 1997b). In the short time ballistic regime the mean squared displacement of grain is $\langle \Delta z(t)^2 \rangle = 2\langle \xi^2 \rangle t^2/\pi\tau_c^2$ and $\langle \Delta z(t)^2 \rangle = 2Dt$ at long times.

The fits of empiric formula to the experimental data are shown in Fig.2and Fig.3. Air blow by pressure of $0.25bar$ gives the best fitting parameters as $\langle \xi^2 \rangle = 2.5\dot{1}0^{-10}m^2$, $\tau_c = 0.22ms$ and $D = 0.93\dot{:}10^{-6}m^2/s$ with the error of about 5%. Strong attenuation at the air blow of $0.5bar$ gives no any evidence about the diffusion constant but very clear exponential dependence with $\langle \xi^2 \rangle = 10.0\dot{1}0^{-10}m^2$, and $\tau_c = 0.33ms$ as shown in Fig.2.

Since the granular temperature is proportional to the averaged grain velocity $T \approx \langle v_z^2 \rangle$, we get increase of temperature for about 60% with the increase of air blow from the pressure from 0.25 bar to 0.5 bar.

4 CONCLUSIONS

The results of our measurement agrees with the results of simulations of hard-sphere fluids of Alder and Wainwright (Alder and Wainwright 1967), who first found that the velocity autocorrelation decays exponentially only for very low densities. If the density of the system is increased the exponential form breaks down and the velocity autocorrelation can become negative with the long range tail due to the caging of particles by their neighbors. They also similar to the measurement by positron emission method in Ref.(Wildman et al. 2002), where the spectrum was explained by the constraining nature of the experimental cell and not by the grain inter-caging as proven by our measurements.

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