Powder characterization by scanning SHG intensity measurements

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We demonstrate an alternative method to characterize the properties of powders based on measuring the polarization dependent second harmonic generation when scanning a focused laser beam across the surface of the powder.

Introduction

Although there are many methods to measure the size distribution of powders, such as microscopy [1] or dynamic light scattering [2], these existing methods suffer from numerous disadvantages when applied in industry during processing (such as milling, granulation, mixing and compaction). Microscopy and dynamic light scattering both require sampling. Dynamic light scattering requires that the particles be diluted and suspended in a non-solvating fluid, while microscopy requires image analysis software to acquire particle sizes from irregularly shaped particles.

In this paper, we demonstrate a new particle sizing method, based on measuring the intensity of optical second harmonic generation (SHG) from individual grains in a powder. Also, unlike earlier measurements [3], we show that the grain size can be obtained independently of the absolute SHG power by analyzing the polarization dependence of SHG when scanning the incident laser beam across the powder surface. This makes the method more robust and thereby meets an important requirement for inline, real-time particle sizing.

Experimental conditions

As shown in Fig. 1, the basis of the experimental setup is a Kerr lens modelocked Ti:Sapphire laser (KM Labs Model TS Ti:Sapphire laser kit, 800 nm, 50 fs, 300 mW, 80 MHz). The linear polarized laser beam is passed through a half-wave plate to set the polarization orientation. To ensure that the laser beam is mostly incident on a single grain in the sample, it is focused to a 20 μm (FWHM) spotsize by a lens (f = 80 mm). The sample is held in a plastic cuvette and is linearly translated up to 6 mm at a speed of 50 μm/s orthogonally to the angle of incidence of the laser beam. The scattered light is collected by an aspheric lens (f = 40 mm, NA = 0.55, 50 mm dia.) placed 3 cm from the point where the laser beam is incident on the sample. The collimated output from the aspheric lens is focused by another lens (f = 100 mm) so that all the collected light is incident on the detectors. The infrared and its second harmonic are separated by a dichroic mirror (Thorlabs FM04). The infrared is detected by a photodiode (BPW 34B), while the second harmonic passes through a bandpass filter (T = 30% 390-400 nm) and is detected by a photomultiplier tube (PMT) (Hamamatsu H6613). The signals from the PD and PMT are recorded simultaneously by an oscilloscope with identical low-pass RC filters (R = 1 kΩ, C = 1 μF). A complete measurement run consists of measuring the fundamental and SHG power as a function of position for ten different polarizations between 0 and 180 degrees.

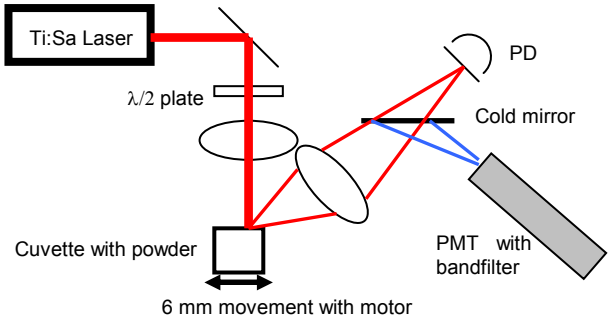

***Fig. 1**: Overview of the experimental setup.*

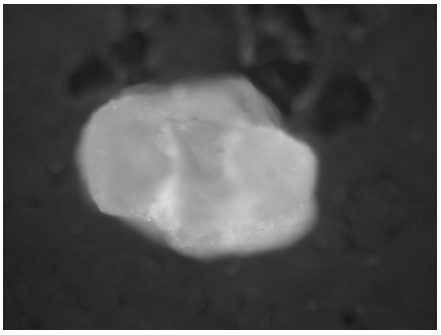

***Fig. 2**: Microscope picture of a single grain of α-lactose with a longest-axis width of 400 µm.*

The samples consisted of α-lactose powder (obtained from the Faculty of Pharmacy, Helsinki University), sieved (Endecotts 150, 106, 72) into five particle size ranges: < 45 µm, 45-71 µm, 71-105 µm, 105-150 µm, and 150-180 µm. In addition, measurements on a single grain (shown in Fig. 2), affixed to a piece of transparent tape, were also carried out.

Results

Fig. 3a shows the power of the SHG and fundamental for a single grain, summed over all polarizations and normalized. The width of the peak of the fundamental light is 500 µm, which is about 25% larger than the grain size, as revealed by microscopy. In contrast, the SHG signal shows two large peaks rather than one. Since such a signal could be erroneously interpreted as two small grains, rather than a single large grain, we investigated the polarization dependence to determine the extent of the grains independently of the SHG power.

Indeed, the measurements showed that the SHG power from a single grain varies sinusoidally with the polarization angle, however, the absolute angle at which the maximum SHG is obtained depends on the relative angle of the grain *and* the polarization. By measuring the SHG power as a function of polarization, the sinusoid can be extracted (by fitting) and the grain boundaries can be found by searching for phase-slips. This is illustrated in Fig. 3b, where the phase of the fitted sinusoid is plotted as a function of position. Note that the phase is constant over the entire range between both SHG power peaks, indicating that this is indeed a single grain. Furthermore, the absolute distance over which the phase remains constant is 400 µm, in excellent agreement with microscope results.

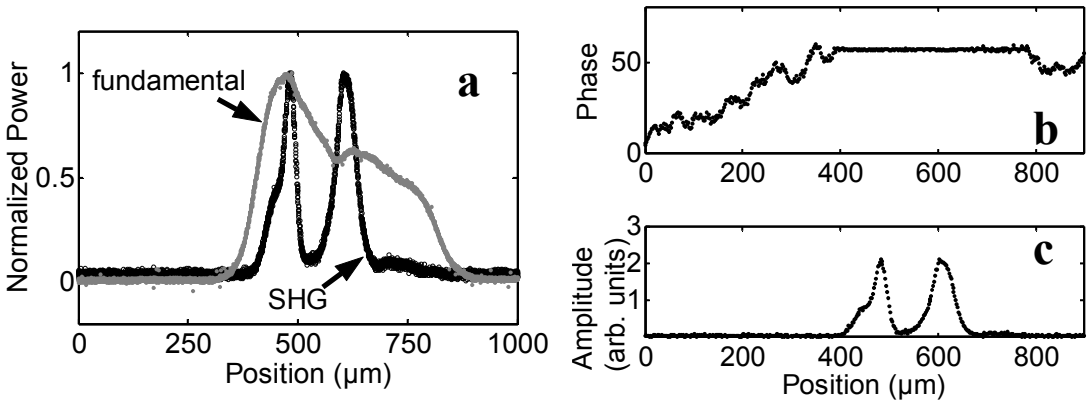

***Fig 3a**: Recorded fundamental (gray) and SHG (black) powers as a function of position across a single grain of α-lactose, summed over all polarizations and normalized. **3b**. Fitted phase (in rad.) of the sinusoid for the SHG power's polarization dependence as a function of position. **3c**. Amplitude of the fitted sinusoid.*

This technique can also be applied to powdered samples. Fig. 4a shows the scattered pump light and SHG power fluctuations as a function of position over a 2 mm scan across powder samples with < 45 μm and 71-105 μm size ranges. Although there are features in these traces, it is difficult to relate the power fluctuations back to a particle size distribution because one must either fit functions, such as a Gaussian, to the peaks or set a threshold, and there is no objective way to distinguish between one large irregular particle and two smaller ones that are partially stacked upon each other.

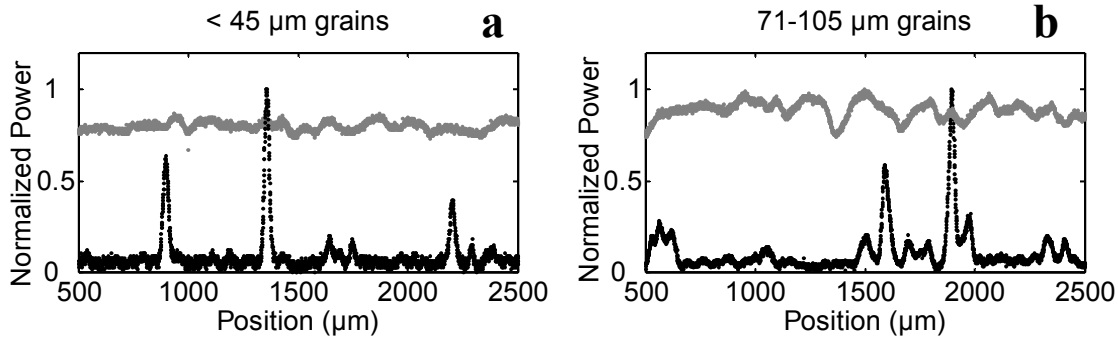

***Fig. 4a**: Fundamental light (grey) and SHG (black) power as a function of position for powders with grains < 45 μm (a) and 71-105 μm (b).*

On the other hand, the polarization dependence shows a distinct sinsusoidal pattern with phase dislocations as a function of position (see Fig 5a). Analysis of the distance between phase dislocations reveals the boundaries of each grain in the scan, as shown in Fig 5b. A histogram of the resulting particle sizes is shown in Fig. 5c, indicating that the polarization dependent SHG is a good mechanism for measuring particle size. Fig. 5d compares the average particle size distribution with that obtained from microscopy. We found that the particle size ranges agree well with each other, though they differ substantially from the range one might expect from the sieve mesh sizes.

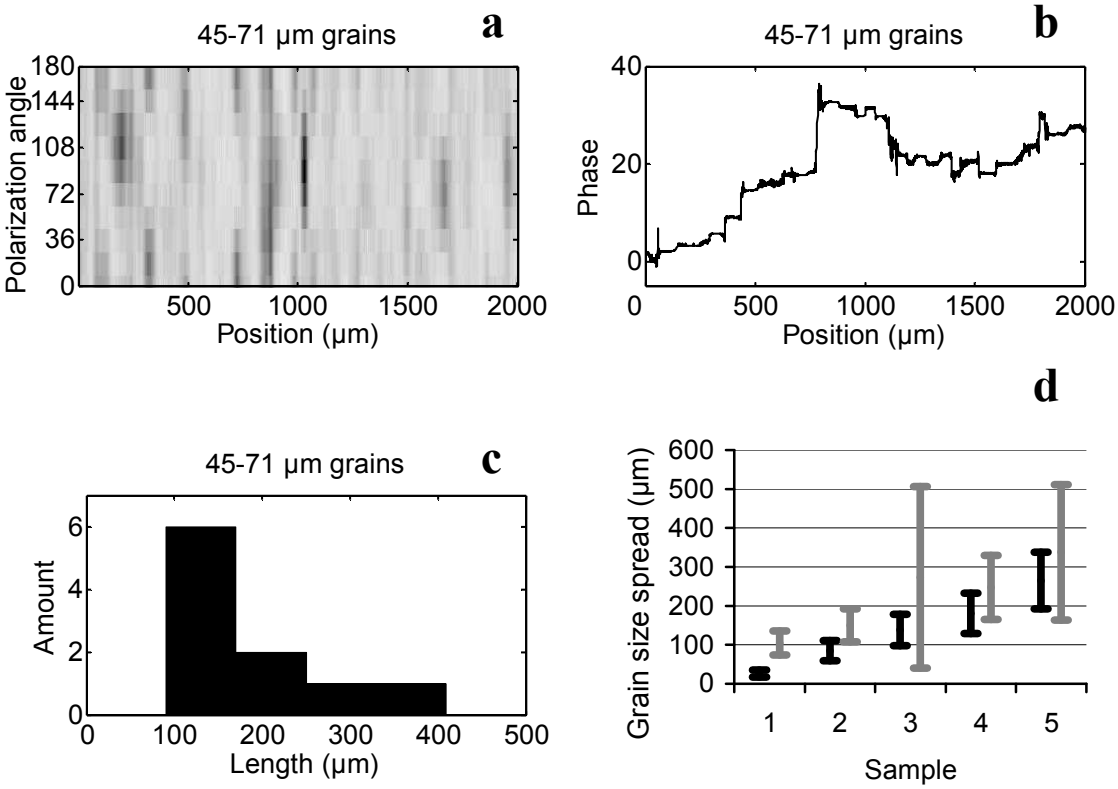

***Fig 5**: SHG power as a function of polarization and position (a). Polarization phase as a function of position (b). Histogram of distance between phase locations (c) for 45-71 µm particles. Comparison of size ranges as measured by microscopy (black) and polarization dependent SHG (grey)(d).*

Conclusions

We have demonstrated a new approach to measure the size of particles in powders via polarization sensitive recordings of the generated second harmonic when scanning a focused laser beam across the powder sample. The simplicity of this method might be interesting for in-line monitoring in industrial applications.

Acknowledgements

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References

[1] N. Laitinen, O. Antikainen and J. Yliruusi, "Does a powder surface contain all necessary information for particle size distribution analysis?", European Journal of Pharmaceutical Sciences, vol. 17, pp. 217-227, 2002.

[2] B.J. Berne and R. Pecora, Dynamic Light Scattering, John Wiley & Sons, 1976, ch. 4, pp. 38-52.

[3] C.B. Rawle, C.J. Lee, C.J. Strachan, K. Payne, P.J. Manson, and T. Rades, Journal of Pharmaceutical Sciences vol. **95**(4), pp. 761-768, 2006.