Light Scattering Device for Measuring Finest Particles in the Exhaust of Diesel Engines

Harald Axmann[1] and Bernd Eichberger[2]
[1]*AVL DiTEST Fahrzeugdiagnose GmbH, Alte Poststraße 152, 8020 Graz, Austria*
[2]*Institute of Electronics, University of Technology, Inffeldgasse 12, 8010 Graz, Austria*

Keywords: Light Scattering, Periodical Technical Inspection, Diesel Engine, Particle Emissions.

Abstract: Recent developments in engine technologies and exhaust aftertreatment systems significantly reduced the particle emissions of diesel engines. This also demands new measurement devices for the periodical emission checks, which shall ensure unchanged low emissions over the vehicles' lifetime. As the current light transmission technique has reached its detection limit, a new device based on light scattering is presented. This paper gives a short overview of scattering theory, followed by a description of the measurement system. An emphasis is placed on the control mechanism for achieving a stable light source. Furthermore first measurement results are presented. Finally the issue of correlation between scattering and established measures is discussed.

1 INTRODUCTION

Particle emissions of internal-combustion engines are a severe problem for health and environment (Mollenhauer and Tschöke, 2007; Nickel et al., 2013; Ristovski et al., 2012). Just recently a study of the World Health Organization found diesel soot to be cancerogenic (IARC, World Health Organization, 2012). In road traffic, particles are mainly produced by diesel engines due to incomplete combustion. Many governments have reacted by introducing limits for the allowed exhaust emissions of newly built vehicles. In Europe these are the European Emission Standards, which have been tightened repeatedly since their creation in 1992. In other countries like Japan or the USA similar regulations have been established (Mollenhauer and Tschöke, 2007). To fulfill the requirements new motor technologies and above all completely new aftertreatment systems like the diesel particle filter (DPF) have been developed (Mamakos et al., 2013; Swanson et al., 2013).

To ensure the compliance of the vehicles with those limits over their lifetime they have to undergo periodic emission checks. In the European Union these are regulated by the directive 2010/48/EU (European Commission, 2010). As an indication of the particle emissions the opacity of the exhaust fumes at free acceleration of the engine to cut-off speed is used. The according measurement device is the opacity meter, which measures the turbidity by means of light transmission. For old diesel vehicles, known to emit black clouds of soot, this method was by far satisfactory. Modern vehicle generations have exhaust gases which are effectively invisible, so this technique reaches its detection limit (Giechaskiel et al., 2013). Due to the poor resolution of the device and the wide measurement tolerances, the limits at the periodic emission checks cannot be tightened. Elevated emissions resulting from damaged DPFs are not detected with the current measurement device, or the cars still pass the check, because the limits are too loose (Boulter et al., 2011; German Association of manufacturers and importers of Automobile Service Equipment, 2010; VdTÜV and DEKRA, 2010).

Consequently new devices for measuring the particle emissions in garages and vehicle inspection institutions are necessary. While providing the necessary sensitivity they also need to fulfill the requirements for the usage in the garages. These are for example simple operation and maintenance, mobility and robustness. In a comparison of different methods for particle measurement another optical principle showed up as most promising: light scattering. Instead of quantifying the attenuation of the transmitted light the portion scattered into a distinct direction is measured (see figure 1). By use of optimized signal processing this method can achieve a sensitivity which surpasses that of opacity meters by two orders of magnitude. The theory behind light scattering shall be explained briefly in the following chapter.

Axmann H. and Eichberger B..
Light Scattering Device for Measuring Finest Particles in the Exhaust of Diesel Engines.
DOI: 10.5220/0004490704570462
In *Proceedings of the 10th International Conference on Informatics in Control, Automation and Robotics* (ICINCO-2013), pages 457-462
ISBN: 978-989-8565-70-9

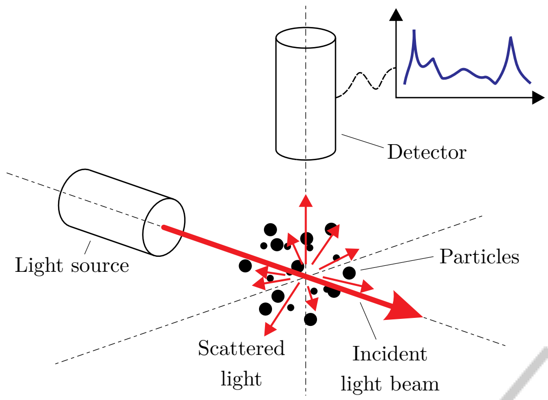


Figure 1: Light scattering principle.

2 LIGHT SCATTERING THEORY

Light scattering is the deflection of light rays due to heterogeneities in the lit medium. The first models for describing the effect of light scattering go back to the 19$^{\text{th}}$ century. The simplest of these models was developed by Lord Rayleigh and deals with very small particles compared to the wavelength of the incident light. This so called Rayleigh scattering produces the blue color of the sky (Bohren and Huffman, 1998; van de Hulst, 1981). For such small particles the scattered light only depends on their size in relation to the wavelength, the polarization, the scattering angle and the optical properties (the scattering index) of the particle. The shape of the particles does not play any role here. The upper limit for the application of the Rayleigh model is reached for particle sizes between one sixth and one tenth of the wavelength.

For bigger particles the shape has an influence on the scattering behavior, too. For the simplest geometric object, the sphere, a complete description of the scattering properties was found in 1908 by Gustav Mie (Mie, 1908). The mathematic functions used herein are by far more complex than the Rayleigh formulas and cannot be calculated in closed form. For small particles both give the same results. Further mathematical models exist for some selected geometric objects like cubes or cylinders, for which the orientation adds as a variable.

For an ensemble of particles with arbitrary shape a point is reached, where exact calculations become too complex and therefore impractical. As the particles are typically randomly oriented, the approach via approximation by spheres can be found in the literature (Görner et al., 1995; Hull et al., 2004). For a random distribution of the particles in space, interference can be neglected and the total scattering can be calculated as the sum of scattering from spherical objects with adequate equivalent diameters. However the shape of soot particles differs considerably from spheres. They are chain-like aggregates of almost spherical monomers, the so-called primary particles (see figure 2). Hence the approximation via spheres must be regarded critically. Although there are methods to exactly calculate the scattering of such aggregates with known structure, this becomes impossible for many particles with various structures and orientations (Chakrabarty et al., 2007). Forrest and Witten found a simpler mathematical description using fractal theory based on the work of Mandelbrot (Forrest and Witten, 1979). The scattering of these fractal-like aggregates has been investigated by Sorensen in extensive studies (Sorensen, 2001). The following central equation relates the number of primary particles N to the geometric aggregate size R_g:

$$N = k_0\,(R_g/a)^{D_f} \tag{1}$$

R_g is a root-mean-square radius called radius of gyration, a is the radius of the primary particles, k_0 a proportionality constant in the order of 1 and D_f the fractal dimension. The latter is a measure of the space filling capacity of objects. Small values mean sparse long chains, whereas values near 3 mean dense sphere-like structures. Soot particles typically have $D_f \approx 1.78$ or $D_f \approx 1.95$, depending on the creation process (Lapuerta et al., 2006).

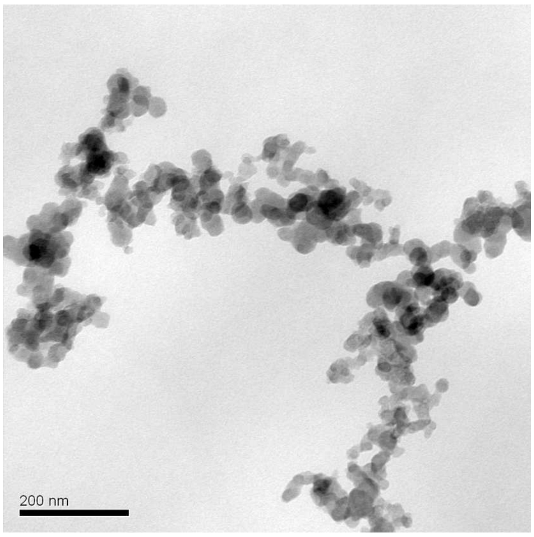


Figure 2: Soot particle from a propane flame under a transmission electron microscope (Reinisch, 2009).

On the basis of equation (1) a structure factor can be calculated for the particle, which describes the interference between the scattering contributions of the primary particles. It is assumed that all monomers are of the same size and see the same incident light wave. Then the total scattering is the sum of the N

single scattering contributions of the monomers multiplied by the structure factor. The scattered light of an ensemble of particles can again be calculated by summation, because of the random distribution of the particles.

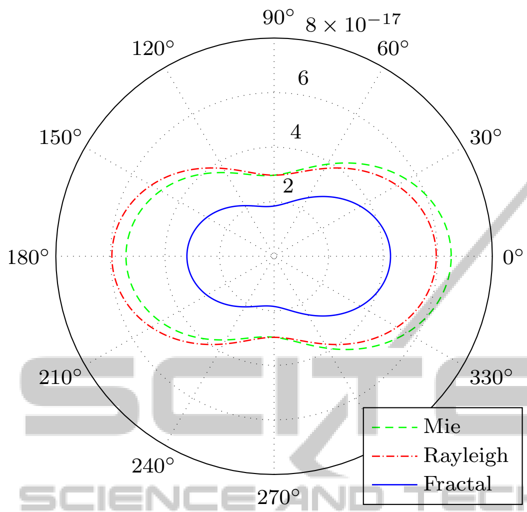


Figure 3: Calculated scattering intensity for a 100 nm particle using various mathematical models.

Figure 3 shows the radial scattering intensity in $W/(m^2 sr)$ of a typical soot aggregate from combustion processes using different mathematical models. The size of the particle (diameter or $2R_g$ respectively) is 100 nm, its complex scattering index is assumed as $1.5 + 1i$. The incident light ray with wavelength 660 nm and an irradiance of $1\,W/m^2$ is directed along the positive abscissa. In the diagram one can nicely see the starting deviation of the scattering between Rayleigh and Mie model. For this size there is just a little bit more forward scattering (i. e. in direction of 0°). Forward scattering will become more and more dominant for bigger particles. Furthermore ripples from interferences appear. In comparison the scattering intensity of the fractal is smaller by two thirds, because it is practically just the sum of the disproportionately weaker scattering of all the small primary particles. In contrast to the Mie model there are no ripples for any particle size. This is due to the fact that the fractal model is based on the mean of the various possible structures and orientations of a fractal-like aggregate with the given parameters.

3 TECHNICAL REALISATION

Light scattering devices are used in non-exhaust related applications, e. g. for air quality monitoring and for basic research (Görner et al., 1995; Hull et al., 2004). However, for the use in the periodic technical inspection special requirements have to be fulfilled. These are robustness, resistance against the sometimes tough environmental conditions in the garages (temperature, humidity), mobility, high dynamic range (suitable for both low and high emitters), low maintenance, short preparation time (e. g. heating up), compatibility with currently used measuring units and finally low costs, to be affordable for the garages.

Light scattering has an essential disadvantage: The intensity of the scattered light is very small compared to the incident intensity. Even if a wide angle area is covered the ratio can be $1:10^9$ or more. Hence strong light sources combined with sensitive detectors are necessary to achieve the desired resolution. Such components, especially strong lasers, are typically rather expensive. In comparison to light from other sources laser light has the advantage to be very well focusable, which is helpful for reducing the stray light. Theoretically in light scattering the sensitivity only depends on the absolute intensity of the incident light, as there should ideally be no scattered light in the absence of particles. In practice stray light and reflections from the interior of the measurement cell will produce some background light. That means that the stability of the light source will directly influence the lower detection limit, too. Noise or drift of the light intensity degrade the performance considerably, and increasing the intensity does not have a positive effect (see figure 4). Therefore the control of the light source is a crucial issue.

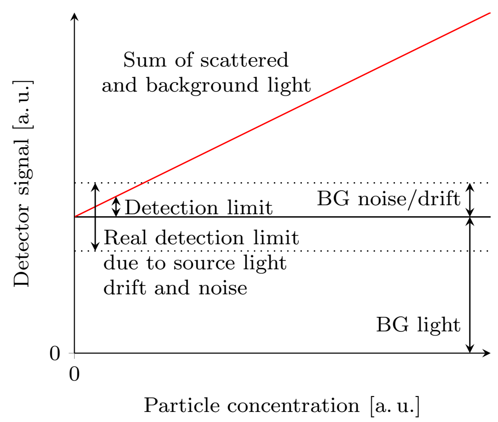


Figure 4: Degraded detection limit due to noise and drift of the background (BG) light in arbitrary units (a. u.).

For diode lasers the intensity is controlled via the current through the diode. There are two principle ways to operate them (Webb and Jones, 2004):

1. Automatic Current Control (ACC) operates the diode with a constant current which is known to

provide the desired intensity.

2. Automatic Power Control (APC) uses a monitor diode to measure the laser optical output power and to adjust the current accordingly.

APC is a simple solution to produce a more or less constant intensity, which is ideally independent of the ambient temperature. However depending on the current and the temperature mode hopping may occur. Mode hopping is the abrupt switching from one longitudinal mode in the laser to another (Heumier and Carlsten, 1993; Pralgauskaitė et al., 2013). It leads to a small change in wavelength and intensity and can even cause variations in the direction of the laser light. Best stability can be achieved with ACC and precise temperature control. Though, for a bad combination of case temperature and current, mode hopping may still occur (Ascente, 2007). Furthermore precise temperature stabilization is costly, so one might not want to integrate it into a garage device.

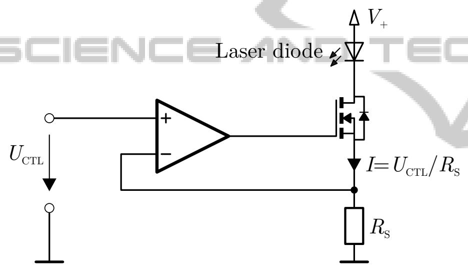
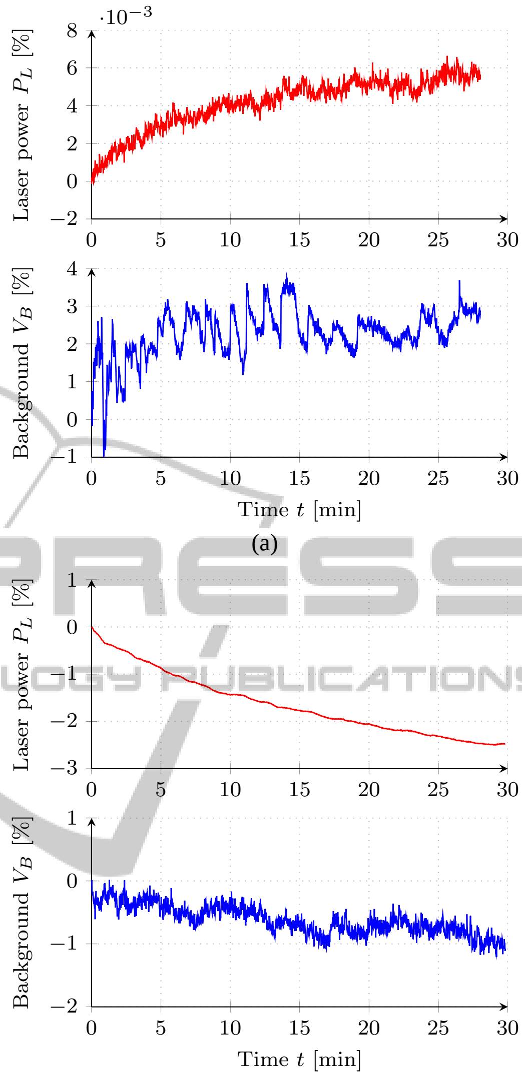


Figure 5: Simplified analog ACC control loop without stability measures.

Several test runs showed that the best results in terms of background light stability can be achieved when using ACC with a constant average current and an AC modulation superimposed (see figure 6). A frequency of a few MHz and a modulation amplitude of 50 % are a good choice. Due to warming of the laser diode in continuous operation the laser intensity decreases with constant current. This leads to a smaller scattering signal and a drop in the signal background and has to be corrected by postprocessing. The effective laser power can be determined via the monitor diode. This information can be used in the micro processor to correct both the scattering and the background signal accordingly. As the stray light might not be directly proportional to the intensity in the focus, zero correction might not work optimally. Still sufficiently stable background light levels could be achieved.

For the actual system red diode lasers with a wavelength of 660 nm and optical powers of 5-15 mW are used. The wavelength is relatively big in comparison to the exhaust particles. The accumulation mode, which contains most of the particle mass, has



(b)

Figure 6: Normalized laser power and background signal in the measurement cell for different laser operation modes without temperature control. (a) APC: The laser power is held almost perfectly constant, the background signal shows drift and spontaneous changes due to mode hopping though. (b) ACC with modulation: The background signal is much smoother and its drift is very small.

its peak around 100 nm (Kittelson, 1998; Liu et al., 2012). This is already near the Rayleigh limit at this wavelength. For particles below this size limit scattering will decrease with the sixth power of the size, quickly reaching the detection limit. Lasers with shorter wavelength are still very expensive. The gain in scattering is furthermore partly suspended by the low spectral efficiency of photo diodes for these wavelengths. Since the soot particles have a very broad size distribution there are still many particles that can be nicely detected using red light.

For detection a small photodiode with 1 mm^2 of sensitive area is used. It is thermally isolated from the measurement cell to reduce temperature influences. The current generated by the photo diode is in the picoampere range and below. It is amplified in a sophisticated circuit using special shielding measures to reduce electromagnetic interferences. Precision analog digital converters typically feature a differential input structure. The single ended current signal of the diode is converted to such a differential voltage as exactly as possible. For this purpose a new amplifier circuit design is used (Axmann and Eichberger, 2012, European patent pending). The photo diode is connected to two symmetric transimpedance amplifiers leading to a differential signal around a center voltage. Noise on the supply line will merely influence this center voltage resulting in a common mode interference, which is suppressed by the analog digital converter.

In combination with a high amplification oversampling is used to achieve a higher resolution. The analog signal is sampled at a rate of 1 MHz, whereas the data output rate is 100 Hz. Test measurements confirmed that this temporal resolution is sufficient to completely trace the exhaust emissions of combustion engines. It is noteworthy that in the measurement circuit a dynamic range of 120 dB is attained with only one amplifier stage. The lowest measurable currents are approximately 250 fA, which is equivalent to approximately 500 fW of optical power.

4 CORRELATION TO ESTABLISHED MEASURES

Measurements confirmed that the new device is comparable to much more expensive test bench equipment in terms of sensitivity. Thus it is capable of reliably detecting smallest defects in the DPF. However, for type approval of the new measurement technique a conversion of the scattering signal to established measures is necessary. For Germany this is the opacity N in percent or the equivalent absorption coefficient k in 1/m. Such a conversion is not straight forward, because there is no direct physical relation between scattering and opacity. A look at the mathematic formulas illustrates that properties like used wavelength or shape, size and optical properties of the particles have an essential impact on the result.

Moreover the measurement principles differ in their sensitivities. For example light transmission is notably sensitive to NO_2. This is a brown gaseous exhaust component, which also attenuates the incident light (Giechaskiel et al., 2013; Mollenhauer and Tschöke, 2007; Norris, 2005). A series of measurements at DPF equipped vehicles revealed that opacity from NO_2 can even be the major part. For light scattering there is no such influence because the gas molecules are below the detection limit. Accordingly light scattering is a pure particle measurement technique. With this in mind a conversion of the scattering signal to a particle mass concentration in mg/m^3 seems reasonable. Photo acoustic soot sensors have proven to be a good reference for real time measurements of the mass concentration. They are based on the resonant measurement of acoustic waves created by periodically absorbing particles. The measured signal is proportional to the soot mass concentration with minimal cross sensitivity (Mollenhauer and Tschöke, 2007). The limitation to soot however makes direct comparison to scattering signals sometimes difficult, e. g. when abrasion particles are involved.

Those considerations lead to the conclusion that a fundamental conversion of the scattering signal to the said measures is impossible. Instead one has to rely on empirical formulas. A series of studies, e. g. (VdTÜV and DEKRA, 2010), and measurements demonstrated that there is a good overall comparability between scattering, opacity and mass concentration. In figure 7 the comparison of a device based on scattering to an opacity meter for soot particles from propane combustion is shown. A good correlation exists in the depicted range. Based on the used conversion formulas the detection limit of light scattering can be derived. It is below $k = 0.0001$ /m. This is an improvement of two orders of magnitude compared to currently available garage opacity meters (Norris, 2005).

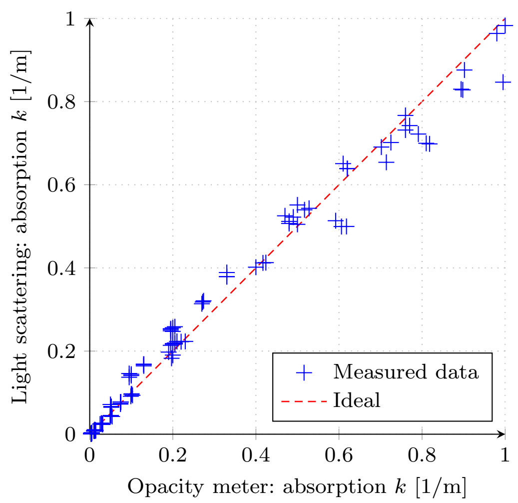


Figure 7: Correlation of the absorption coefficient between an opacity meter and a light scattering device ($k \leq 1$ /m).

5 CONCLUSIONS

A particle measurement method for periodic emission tests based on light scattering has been presented. Care must be taken that noise from the incident light (e. g. due to mode hopping for laser sources) is minimized to achieve highest possible resolutions. In combination with a sensitive detection system resolutions 100 times better than that of current garage opacity meters can be achieved at comparable costs. The temporal resolution (100 Hz) is by far sufficient for tracing exhaust emissions. Thus light scattering devices are suitable for the exhaust measurement of vehicles with state-of-the-art aftertreatment systems. Further work will focus on the conversion of these results to established measures like opacity in order to ensure the applicability of the determined formulas.

REFERENCES

Ascente, T. (2007). Laser diode intensity noise induced by mode hopping. *Rom. Rep. Phys.*, 59(1):87–92.

Axmann, H. and Eichberger, B. (2012). Circuit configuration for evaluating photoelectric measurements. European Patent Application EP2501036A1.

Bohren, C. F. and Huffman, D. R. (1998). *Absorption and scattering of light by small particles*. Wiley, New York.

Boulter, P., Buekenhoudt, P., Stricker, P., et al. (2011). TEDDIE – A new roadworthiness emission test for diesel vehicles involving NO, NO_2, and PM measurements. Final report, CITA.

Chakrabarty, R. K., Moosmüller, H., Arnott, W. P., et al. (2007). Light scattering and absorption by fractal-like carbonaceous chain aggregates: comparison of theories and experiment. *Appl. Optics*, 46(28):6990–7006.

European Commission (2010). Commission Directive 2010/48/EU of 5 July 2010 adapting to technical progress Directive 2009/40/EC of the European Parliament and of the Council on roadworthiness tests for motor vehicles and their trailers. OJ L 173:42-72.

Forrest, S. R. and Witten, T. A. (1979). Long-range correlations in smoke-particle aggregates. *J. Phys. A: Math. Gen.*, 12(5):L109–L117.

German Association of manufacturers and importers of Automobile Service Equipment (2010). Diesel-AU: Die Endrohrprüfung ist unerlässlich. Press Info. 4/10.

Giechaskiel, B., Schiefer, E., Schindler, W., Axmann, H., et al. (2013). Overview of soot emission measurements instrumentation: From smoke and filter mass to particle number. *SAE Int. J. Engines*, 6(1):10–22.

Görner, P., Bemer, D., and Fabriés, J. (1995). Photometer measurement of polydisperse aerosols. *J. Aerosol Sci.*, 26(8):1281–1302.

Heumier, T. A. and Carlsten, J. L. (1993). Detecting mode hopping in semiconductor lasers by monitoring intensity noise. *IEEE J. Quantum Elect.*, 29(11):2756–2761.

Hull, P., Shepherd, I., and Hunt, A. (2004). Modeling light scattering from diesel soot particles. *Appl. Opt.*, 43(17):3433–3441.

IARC, World Health Organization (2012). Diesel engine exhaust carcinogenic. Press Release No. 213.

Kittelson, D. B. (1998). Engines and nanoparticles: A review. *J. Aerosol Sci.*, 29(5/6):575–588.

Lapuerta, M., Ballesteros, R., and Martos, F. (2006). A method to determine the fractal dimension of diesel soot agglomerates. *J. Colloid Interface Sci.*, 303:149–158.

Liu, Z., Swanson, J., Kittelson, D. B., and Pui, D. Y. H. (2012). Comparison of methods for online measurement of diesel particulate matter. *Environ. Sci. Technol.*, 46(11):6127–6133.

Mamakos, A., Martini, G., and Manfredi, U. (2013). Assessment of the legislated particle number measurement procedure for a euro 5 and a euro 6 compliant diesel passenger cars under regulated and unregulated conditions. *J. Aerosol Sci.*, 55:31–47.

Mie, G. (1908). Beiträge zur Optik trüber Medien, speziell kolloidaler Metallösungen. *Ann. Phys.*, 25(3):377–445.

Mollenhauer, K. and Tschöke, H. (2007). *Handbuch Dieselmotoren*. Springer, Berlin Heidelberg, 3rd edition.

Nickel, C., Kaminski, H., Hellack, B., Quass, U., et al. (2013). Size resolved particle number emission factors of motorway traffic differentiated between heavy and light duty vehicles. *Aerosol. Air Qual. Res.*, 13(2):450–461.

Norris, J. O. W. (2005). Low emission diesel research CP17/18/770. Phase 3 – Report, Vehicle and Operator Services Agency (VOSA).

Pralgauskaitė, S., Palenskis, V., Matukas, J., et al. (2013). Analysis of mode-hopping effect in Fabry-Pérot multiple-quantum well laser diodes via low frequency noise investigation. *Solid State Electron.*, 79:104–110.

Reinisch, T. (2009). Über die Morphologie von Rußpartikeln von Verbrennungskraftmotoren im Hinblick auf Partikelanzahlmessung. Bachelor Thesis, Graz, University of Technology.

Ristovski, Z. D., Miljevic, B., Surawski, N. C., Morawska, L., et al. (2012). Respiratory health effects of diesel particulate matter. *Respirology*, 17(2):201–212.

Sorensen, C. (2001). Light scattering by fractal aggregates: A review. *Aerosol Sci. Technol.*, 35(2):648–687.

Swanson, J. J., Watts, W. F., Newman, R. A., et al. (2013). Simultaneous reduction of particulate matter and NO_x emissions using 4-way catalyzed filtration systems. *Environ. Sci. Technol.*, 47(9):4521–4527.

van de Hulst, H. C. (1981). *Light scattering by small particles*. Dover Publications, New York.

VdTÜV and DEKRA (2010). Emission Check 2010 Teil 2 – Diesel.

Webb, C. E. and Jones, J. D. C., editors (2004). *Handbook of Laser Technology and Applications*, volume I: Principles. Institute of Physics Publishing, Bristol, Philadelphia.