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Laser Cleaning

Dr. Andrew C. Tam

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Preface

Within the catastrophe theory there is a mathematical statement known as the “Principle of Fragility of Good Things” (Arnold, 1992). According to this principle the stability domain on the plane of parameters (on which the equilibrium state depends) is arranged “with corners outward”, thus driving wedges into the instability domain. As Arnold wrote: “Thus for systems belonging to the singular part of the stability boundary, a small change of the parameters is more likely to send the system into the unstable region than into the stable region. This is a manifestation of a general principle stating that all good things (e.g. stability) are more fragile than bad things”. It is easy to explain this principle from the common sense point of view. To be “good”, regardless of how one defines this word, a number of requirements must hold *simultaneously*, while *even one* failure will suffice for a situation to be called bad.

It is rather difficult to be “good” when working at the frontiers of technologies, such as microelectronics, micromechanics, and data storage. These technologies should meet a range of specifications, which are often in competition with each other. It is surprising that it is possible to satisfy them. The restrictions of high technologies compared to conventional technologies are not related to the skill of the worker or the quality of the equipment. These restrictions are caused by the physical nature of the process (sometimes it is called a “theoretical limit”). High technologies are closely connected with high science and there is a fruitful physical idea at the basis of each technological victory. IBM, Philips and other top R&D Centers are not only famous for their technological achievements but also for their outstanding fundamental research. A textbook example is the research on semiconductors in the Bell Telephone Laboratory as well as the discovery of the transistor effect.

Having in mind the laser cleaning process one should start with the idea of adhesion. Researchers at the Philips research laboratory have contributed considerably to the development of this idea. It is sufficient to mention just two names – Hamaker and Casimir.

In 1937 Hamaker accomplished a very clear presentation of the attraction of particles to a surface by the van der Waals force. Although the corresponding potential varies rapidly with distance, as r^{-6} (Landau & Lifshitz, *Quantum Mechanics*), it represents a long-range interaction. The

contribution of this interaction to the free energy is not additive; it depends on the body shape and configuration. If one considers the particle as a deformed sphere, then, according to Hamaker, the attraction force is given by

$$F = \frac{Aa}{6h^2} \left(1 + \frac{r_c^2}{ah} \right), \quad (1)$$

where a is the radius of the particle; h is the separation distance, which is understood as an equilibrium distance originated by interplay of attraction and repulsion forces ($h \approx 4 \text{ \AA}$); and r_c is the radius of contact. The Hamaker constant $A \approx (0.5 - 5.0) \cdot 10^{-19} \text{ J}$ depends on the properties of the particle, substrate and medium. This value has been measured for many materials. In the absence of additional load, for (sub) micrometer-sized particles the second term in the bracket is the most important, $r_c^2/ah \gg 1$. The characteristic adhesion force for a $1 \text{ }\mu\text{m}$ spherical particle is of the order of 0.1 dynes . It is a big force having in mind that modern techniques permit the measurement of a force a billion times smaller (Rugar, 2001). It is clear that this high loading leads to elastic or even plastic deformation of the material. These deformations define the size of the contact region.

In 1882, Hertz performed the first examination of the pressure distribution in the contact area. This distribution follows a parabolic law:

$$P(r) = P_{\max} \left(1 - \frac{r^2}{r_c^2} \right)^{1/2}, \quad (2)$$

(see, for example, the analysis of the Hertz solution in § 9 in Landau & Lifshitz "Theory of Elasticity"). According to (2), the maximum pressure, $P_{\max}$, is 1.5 times higher than the average pressure in the contact area

$$P_{\max} = \frac{3}{2} \frac{F}{\pi r_c^2}. \quad (3)$$

Assuming a Hertzian pressure distribution, Derjaguin in 1934 found the relation for the radius of contact, r_c , versus particle size:

$$r_c = \left(\frac{Aa^2}{8E^*h^2} \right)^{1/3}, \quad \frac{1}{E^*} = \left(\frac{1-\sigma_p^2}{E_p} + \frac{1-\sigma_s^2}{E_s} \right), \quad (4)$$

where σ and E are the Poisson coefficients and Young's modulus, and subscripts "p" and "s" indicate the particle and the substrate, respectively.

From (1)-(4) one can find the relation:

$$P_{\max} = \frac{A}{4\pi h^3} + \frac{1}{\pi} \left(\frac{E^* A}{h^2 a} \right)^{1/3}. \quad (5)$$

The value of the van der Waals attraction between planes (per unit of area) is given by the first term of (5).

One can see that $P_{\max}$ becomes higher for smaller particles. For particles with a size of the order of 10 nm (nanoclusters) this pressure can reach a value of the order of one hundred kbar.

The adhesion-induced deformations are quite complex, and some other factors, such as adhesion forces outside the area of contact, should be taken into account to describe the experimental data well. At present, two models of adhesion are commonly acceptable: the Derjaguin-Muller-Toropov (DMT) model for "hard" materials (Derjaguin, 1975, 1980; Muller, 1983) and the Johnson-Kendall-Roberts (JKR) model for "soft" materials (Johnson, 1971, 1976). The transition between the two models also has been discussed (Maugis, 1992, 1995; Greenwood, 1998).

One should pay attention to the phenomena of instability, which are important for the understanding of laser cleaning. Namely, the jump-like disconnection of the particle from the surface arises when the critical negative force (tensile) is applied. On the contrary, particles can approach the surface without a load but when the particle touches the surface the jump-like adhesion force arises. This hysteresis phenomenon can be verified experimentally when the load of the particle is performed with the help of an atomic force microscope (AFM). Such experiments are used to measure the Hamaker constants (see, for example, Shaefer, 1995; Mizes, 1995).

The known paradox in the history of science is that sometimes discoveries are attributed to people who did not make these discoveries. For example, the exponential decay of the intensity in absorbing media, $I = I_0 \exp[-\alpha z]$, is frequently called Beer's law. Actually this law was

experimentally established by P. Bouguer in 1729 and was deduced theoretically by J. H. Lambert in 1760. In 1852 A. Beer established another law on the relation of the absorption coefficient to the density N of the absorbing particles, $\alpha = \sigma N$, where σ is the absorption cross-section. It is similar to the association of the name of Christopher Columbus with the discovery of America. Actually, America had been discovered before Columbus. Moreover, Columbus has not even rediscovered America. His caravels had only reached the islands Cuba and Haiti, while we know the continent of America due to Amerigo Vespucci.

Hamaker did not perform any calculations of the Hamaker constant for solids. In his paper he introduced the value $A = \pi q^2 \lambda$, where q is the number density of interacting atoms and λ is a London constant, which describes the dipole-dipole interaction between isolated atoms. This formula can be applied to the description of attraction within a diluted gas but not to solids, where the situation is much more complex.

Casimir carried out the first "real calculation" of the Hamaker constant for solids in 1948. He made the calculations of the attraction force between conducting plates; now this force is called the "Casimir force". This force is universal and it does not depend on the metal properties. The value of the Casimir force (per unit of area) is given by

$$F_{\infty} = \frac{\pi^2}{240} \frac{\hbar c}{h^4}. \quad (6)$$

The Casimir force arises due to quantum vacuum fluctuations of the electromagnetic field. Fluctuations outside the plates are unrestricted, whereas between the plates some restrictions occur due to the boundary conditions. The Casimir force is remarkable, because it is a more palpable consequence of the zero-point field than the Lamb shift (Bordag, 1999; Maclay, 2001). The Casimir force is very big and plays a major role in modern Micro Electro Mechanical Systems (MEMS) (Chan, 2001).

In the framework of macroscopic theory, the van der Waals interaction in a material medium is considered to be caused by a long-wavelength electromagnetic field; Lifshitz suggested this concept in 1955 (see also § 90 in Landau & Lifshitz, *Electrodynamics of Continuous Media*). It includes not only thermal fluctuations but also the zero-point oscillations of the field.

The Lifshitz theory is based on the theory of electromagnetic fluctuations developed by Leontovich (1952) and Rytov (1953). Later, the

Lifshitz formula was derived from the microscopic point of view, using the methods of the quantum theory of the field (Dzyaloshinski, 1959, 1960 a, b; Abrikosov, 1965; see also §§ 80-82 in Lifshitz & Pitaevskii, *Statistical Physics, Part 2*). Josef Brodsky said that with the growth of a poet's skill the number of his readers shrinks. I understand the reason for this very well, because I remember the efforts I made during my studies to reproduce the results on the van der Waals attraction from the book of Abrikosov, Gorkov and Dzyaloshinski. In the static limit, the permittivities ε_i can be replaced by their electrostatic dielectric constants ε_{i0} . Then the formula for the attraction force can be written in simplified form:

$$F = \frac{\hbar c}{32\pi^2 h^4} \int_0^\infty dx \int_1^\infty dp \frac{x^3}{p^2} \left\{ \left[\frac{(s_{10} + p)(s_{20} + p)}{(s_{10} - p)(s_{20} - p)} e^x - 1 \right]^{-1} + \left[\frac{(s_{10} + p\varepsilon_{10})(s_{20} + p\varepsilon_{20})}{(s_{10} - p\varepsilon_{10})(s_{20} - p\varepsilon_{20})} e^x - 1 \right]^{-1} \right\}, \quad (7)$$

$$s_{10} = \sqrt{\varepsilon_{01} - 1 + p^2}, \quad s_{20} = \sqrt{\varepsilon_{02} - 1 + p^2}.$$

It is readily seen that in the limit of two good metals, $\varepsilon_{10} = \varepsilon_{20} \rightarrow \infty$, formula (7) yields the Casimir force (6). This force varies strongly with distance h . For a typical separation distance in MEMS of $h = 10$ nm, the value F_∞ is about 1 bar while for a typical separation thickness in adhesion of $h = 4$ Å, the value F_∞ is about 0.5 megabar.

The Lifshitz theory suggests that the van der Waals interaction between solids arises due to the exchange of electromagnetic fluctuations with characteristic energy $\langle \hbar\omega \rangle$ (this value is called the Lifshitz - van der Waals constant). The Hamaker constant is related to the Lifshitz - van der Waals constant by $A = \frac{3}{4\pi} \langle \hbar\omega \rangle$. The numerical factor $\frac{3}{4\pi}$ has no physical relevance.

A simple formula for metals follows under the approximation of the Drude theory:

$$\langle \hbar\omega \rangle \approx \frac{\pi^{3/2}}{8} \hbar \omega_p, \quad (8)$$

where ω_p is the plasma frequency, and γ is the frequency of collisions, which is considered to be small, $\gamma \ll \omega_p$. For aluminum, the only real Drude metal, formula (8) yields a value $\langle \hbar\omega \rangle = 8.11$ eV, which is close to the value of $\langle \hbar\omega \rangle = 8.59$ eV that follows from the integration of an experimental spectrum.

Klimchitskaya (2000), discussed the generalization of the Lifshitz formalism for the case when the spatial dispersion can be important, in addition to the frequency dependence. Another problem is the feedback caused by the variation of a dielectric constant with pressure. Here I want to draw attention to one possible effect. With higher loading pressure one has a higher ε and a higher attraction force. This positive feedback may lead to bistability in optical parameters (either in the Lifshitz-Hamaker constant, or in the attraction force) versus particle size or loading force. Formally, two curves: $F = F(\varepsilon)$ from (7) and $\varepsilon = \varepsilon(F)$ produce a diagram, similar to Semenov's diagram in the theory of combustion (Karlov, 2000). The bistability looks like an optical phase transition, which is attractive for applications to high-density information recording.

Great scientists like Casimir and those from the Landau school created the basic understanding in adhesion. They say that when Wolfgang Pauli learnt that Casimir had become director of the Philips Research Laboratories, he exclaimed: "Well, now I'll stop reading Casimir's papers, as it is clear - what papers directors write!" Probably this anecdote came from Casimir himself; he enjoyed telling anecdotes about his relationship with Pauli. In fact, Casimir wrote outstanding papers both before and during his directorship' period. He discussed the retardation effects in attraction (see e.g. Landau & Lifshitz, *Quantum Electrodynamics*) and many other perfect physics.

Adhesion by itself is not the only problem, which is necessary for the understanding of laser cleaning. We should mention Casimir's name once again because of the problem related to the diffraction of light by the particle (Bouwkamp & Casimir, 1954). In this paper it was shown that spatially distributed electrical currents could be represented as a series of multipole fields. This allows, for example, the calculation of antennas with arbitrary current distributions. The same method was used to find the exact solution for the problem of "particle on the surface" (Bobbert, 1986). Recently, a new interest in this problem has been raised by the study of near-field effects in laser cleaning (Luk'yanchuk, 2000).

No doubt, great and beautiful physical ideas encourage the advancement of new technologies. However, in the relations of science and technology a feedback, which produces the so-called “science-technology spiral”, plays a very important role; Casimir discussed this problem in 1983. Russian poet Anna Akhmatova wrote: “I wish you were aware from what stray matter springs poetry to prosper without shame...” The soil of science is not nobler than that of poetry. Many problems, which lead to great discoveries started off as purely technological studies. I want to present just two examples.

The first is related to the problem of minimization of scraps of a carton sheet during its cutting solved by L. V. Kantorovich. Kantorovich was a mathematical genius (a Full Professor of mathematics at Leningrad State University at the age of 18). He did a lot of outstanding mathematical discoveries: the Kantorovich space, the Hahn Banach - Kantorovich theorem, etc. (Kantorovich, 1996). But the highest honor given to him is closely associated with the solution of the problem of scrap minimization mentioned earlier. To solve this problem Kantorovich has developed a new mathematical technique of linear programming, which later earned him the Nobel Prize in economics.

Another example is the paper by Landau & Levich, 1942, devoted to dragging of a liquid by a moving plate. This paper was also motivated by a purely technological problem (deposition of a uniform layer of photosensitive emulsion upon the cinefilm base). It is worth mentioning that the entire modern theory of dynamic wetting and interfacial hydrodynamics has come out of this work (see Landau & Lifshitz, Fluid Mechanics; Levich, 1962).

While preparing this book, I had been very interested in the history of laser cleaning. To the best of my knowledge the first papers devoted to laser cleaning of the surface from small particles were published in 1987-88 (Beklemyshev, 1987, 1990, 1991; Assenfel't, 1988 a, b; Petrov, 1989; Balychenko, 1991). One of the co-authors of these papers was late A. M. Prokhorov, a Nobel Prize winner. These experimental results received good theoretical support (Kolomenskii, 1991a, b). Finally, the Russian group obtained a patent for laser cleaning (Boykov, 1991). Unfortunately, Dr. Yu. N. Petrov, who was the “driving force” behind Russian laser cleaning, passed away at the beginning of the 90's. Further development of the concept of laser cleaning was accomplished by Kolomenskii (1993a, b, 1995, 1998); Mikhalevich (1994); Maznev (1995), who presented several

new and interesting effects such as the excitation of nonlinear surface acoustic waves and the phonon focusing effect.

Prof. S. D. Allen is another well-known name in laser cleaning. Her group has been studying laser cleaning since 1987, while the first papers were published in 1991 (Imen, 1991a; Lee, 1991). Her group received an USA Patent on laser cleaning in 1991 (Imen, 1991b). Among the pioneering research results of S. D. Allen are a number of papers (Lee, 1992, 1993a, b).

Last but not least the group of Dr. W. Zapka should be mentioned. Probably Zapka's contribution has been the most important for industrial applications of laser cleaning. In any case, he received the first European patent on laser cleaning in 1989 (priority from 1987). Although it is not possible to restore the history with the accuracy of one day or one week, my examination shows that in 1987 three groups (Yu. Petrov in Russia, S. D. Allen in USA, and W. Zapka in Germany) discovered the laser cleaning effect simultaneously and independently. The achievement of Petrov's group was that they published the first papers on laser cleaning, while Zapka's group obtained the first patent and realized the first industrial applications. Nevertheless if somebody asked me to recommend people for an award for the discovery of laser cleaning, I would mention all the three names.

This book is dedicated to Dr. Andrew C. Tam from IBM Almaden Research Center. He was not the first to discover the laser-cleaning effect. His first publications appeared only in 1991 (Zapka, 1991 a, b). Nevertheless if one examines the citation index of papers on laser cleaning he will find that papers (Zapka, 1991 b) and (Tam, 1992) are the most cited. Other papers of A. C. Tam written in collaboration with the groups of C. P. Grigoropoulos and P. Leiderer (Park, 1992, 1996; Leung, 1992, Yavas, 1993, 1994 a, b) also have a very high citation impact. At present about half of the citations on laser cleaning are related to the papers with A. C. Tam's name. One can find a parallel in the history of science. It is well known that Galileo was not the first to discover the telescope. Nevertheless only after Galileo's demonstration of Jupiter's moons the telescope became popular throughout the whole world. Andrew Tam definitely had Galileo's talent as a popularizer.

Within 5 to 6 years after Tam's investigations scientists from many countries published more than one hundred papers devoted to laser cleaning. It is not possible to recall the history of these research results objectively. Here different storytellers state the same events in a completely different

ways like in the famous novella of Ryunosuke Akutagawa (and in the not less famous film “Rashomon” of Akira Kurosawa).

At this point it is necessary to mention the work of A. C. Engelsberg (1994), which clearly presented the problem with industrial applications of laser-assisted cleaning technology. Papers of J. D. Kelly (1993) gave the foundation for the 1D model of dry laser cleaning. For 10 years this model has been the basic model for the interpretation of dry laser-cleaning experiments.

One can say also a few words about the papers of Y. F. Lu. He started with his research in the group of Prof. M. Takai (Osaka University, Japan). Later Y. F. Lu moved to Singapore where he set up a highly active research group. His group published more than 40 papers, devoted to different aspects of laser cleaning. Probably the most famous paper is related to laser cleaning of magnetic head sliders (Lu, 1996), but other papers (Lu, 1994 a, b, c; 1997a, b; 1998a, b, c; 1999, 2000a, b, c; 2001) are also well known. Papers of Y. F. Lu are the most cited after A. C. Tam's papers.

I remember how Prof. I. Dzyaloshinski told the young PhD students in the Landau Institute: “I am very sorry that you came to physics too late when all the simple problems had been solved by your teachers. Thus you have to solve only very difficult problems!” I would say that the period of easy results in laser cleaning was over around 1995. In Lambda Highlights No. 47 (May 1995) the editor published a perfect review on “Surface cleaning by UV laser radiation becomes economical”. At the end of this review he mentioned the works of Prof. C. Fotakis' group, devoted to laser cleaning of artworks. This paper in Lambda Highlights outlined the end of the first stage in the research on laser cleaning.

A new vortex in laser cleaning was initiated after 1998 due to very intensive work of the network “Laser cleaning” of the European Community (see, e.g. papers: Zafiropulos (1998); Dobler (1999); Mosbacher (1999, 2000); Oltra (2000); Leiderer (2000); Fourier (2001)). This European project coordinates the efforts of 8 Universities and Institutes from 5 European countries.

New and more detailed research works were done by Halfpenny (1999, 2000); Kane (2000); Wu (1999a, b, 2000); G. Veerecke (1999 a, b, 2000) and other groups (for example, She (1999); Kim (2001); Yavas (2000); Feng (1999); Schmidt (1999); Lee (2000, 2001); Cetinkaya (2000); Zheng (2001); Luk'yanchuk (2001, 2002). In addition, D. Bäuerle (2000), presented a comprehensive review on laser cleaning. In Singapore we

decided to organize an International Workshop to discuss new trends in laser cleaning. This Workshop was held in May 2001 within the frame of the 2nd International Symposium on Laser Precision Microfabrication (LPM 2001). Miyamoto (2002) published the proceedings of this Symposium.

Since 1984 I have met Dr. A. C. Tam at several international conferences and was astonished by his fast scientific reaction as well as by his nontrivial physical and technological ideas. I remember how Nikita Arnold, who was at that time a student in my group at the General Physics Institute, tried to reproduce some of Tam's results. Now Nikita presents the complete theory of 1D effects in dry laser cleaning (Chapter 2). When I started to do my own calculations in laser cleaning, Andrew Tam was among the first people with whom I discussed the results. Dr. A. C. Tam actively participated in the organization of the Workshop on Laser Cleaning and we planned his opening talk "*The emergence of laser cleaning for Hi-Tech industrial applications*". Our grief was very deep when we received the sad news about Andrew Tam's death. I am grateful to the specialists of many countries who found it possible to come to the Workshop to give tribute to Andrew Tam. In memory of A. C. Tam's input in science and technology, as well as his great contribution to laser cleaning, we decided to publish this book. It presents the review papers of the participants of the 1st International Workshop on Laser Cleaning. This book reflects recent achievements in laser cleaning and can be used as a modern introduction for master's and PhD students and engineers.

Werner Zapka (Chapter 1) introduces the history of the discovery of laser cleaning and Andrew Tam's role in these studies. The three consequent parts of the book are devoted to dry and steam laser cleaning and laser cleaning of artworks. The last part of the book illustrates some applications of laser cleaning.

Laser cleaning contains a broad range of interesting and numerous physics. It is sufficient to say that within the papers devoted to laser cleaning one can meet references to essentially all the volumes of the Landau & Lifshitz Course on Theoretical Physics. For example, "*Mechanics*" can be mentioned when one discusses oscillations of the particles at dry laser cleaning. "*Statistical Physics, Part I*" and "*Physical Kinetics*" are more cited when one discusses the nucleation and bubble formation at steam laser cleaning.

Molière's monsieur Jourdain had no idea for 40 years that he spoke prose. I did not know either that my life for a long time produced the "Brownian pushes", encouraging me to take a step on the "road to laser cleaning". Many people supported me on my way. I am very thankful to my colleagues in the laser group of DSI (especially to W. D. Song, M. H. Hong and Y. W. Zheng) for the related discussions.

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When preparing this book we have used "internal" reviewing of the papers. In fact, all the authors have helped me with their critical comments. Profs. S. I. Anisimov, D. Bäuerle, P. Dyer, A. Rubenchik, M. Tribelsky and Dr. E. Gatskevich made many useful suggestions as "external" referees. Profs. S. Allen, C. Grigoropoulos, S. Mikhalevich, M. Takai, H. Coufal and Drs. A. Kolomenskii and V. Pustovoy all sent me their comments on the history and the first stage in laser cleaning.

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