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IS&T, *Journal of Imaging Science and Technology*,
7003 Kilworth Lane, Springfield, VA 22151 USA

Pamela J. Forness
Program Manager and Managing Editor of *JIST*:
The Journal of Imaging Science and Technology
IS&T: The Society for Imaging Science and Technology
7003 Kilworth Lane
Springfield, Virginia USA 22151
Voice: 703-642-9090 extension 16
FAX: 703-642-9094
Email: pam@imaging.org <<mailto:pam@imaging.org>>
WEB: <http://www.imaging.org>

Proximity Theory of Toner Adhesion

L. B. Schein*[†] and W. Stanley Czarnecki[†]

Aetas Technology Incorporated, Irvine, California, USA

Measurements of toner adhesion are usually at least one order of magnitude larger than predicted by image force calculations that model the toner charge by locating it in the center of the toner particle. In order to account for this discrepancy, it has been suggested that either the toner charge is not uniformly distributed on the surface or that van der Waals forces dominate toner adhesion. We propose an alternative model of toner adhesion, which takes into account a recent result that shows that there is an electrostatic proximity force of adhesion at the contact point between a toner particle and a conductive plane.

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Introduction

Toner adhesion is active in all of the steps of electrophotography in which toner is moved between surfaces, including development, transfer, and clean. Yet the source of toner adhesion has not been agreed upon, despite intensive investigations starting with the early work of Goel and Spencer,¹ and continuing with a recent review by Hays,² and papers by Gady and coworkers.³ Measurements^{1–7} generally indicate that the force of adhesion is at least one order of magnitude larger than expected by simple image force calculations that model the toner charge by locating it in the center of toner particle. In order to account for this large discrepancy, Hays² proposed that one must take into account non-uniform surface charge distributions and Gady and coworkers³ applied classical JKR theory to calculate the surface adhesive van der Waals forces. Published toner adhesion measurements and these theories are reviewed below.

In this article we propose an alternative theory of toner adhesion. It uses the recently discovered result that a spherically symmetric distribution of charge points cannot be modeled as having its charge in its center unless it is in free space. In contact with a conductive ground plane, there is an additional contribution to the force of adhesion, which has been given the name proximity force, which equals $4/\pi$ times the usually assumed image force.⁸

In this new theory of toner adhesion, we suggest that at every contact point this proximity force is active. Therefore the force of adhesion has three terms, the usually assumed image force, the proximity force times the number of contact points, and the van der Waals force times the number of contact points. This new theory is discussed and our results are summarized in the present article. Experimental verification is described in the following article.

Measurements of Toner Adhesion

There have been many measurements of toner adhesion, beginning with the work of Goel and Spencer.¹ Techniques include the use of centrifuge and Coulomb forces. All of the published measurements indicate very large values compared to simple theory, and very wide distributions.

As an illustration we quote results from Hays' review.² Tables I and II show data taken from the literature by Hays. Table I shows data from centrifuge measurements; Table II shows data taken from electric field detachment experiments. Columns 2–4 characterize the toner particle (diameter, charge-to-mass ratio, and charge-to-diameter ratio). The fifth column quotes the measured adhesion and the sixth column is the calculated adhesion based on the simple image force calculation:

$$F_i = \frac{\alpha}{4\pi\epsilon_0} \frac{Q^2}{d^2} \quad (1)$$

where Q is the toner charge, d is the toner diameter, ϵ_0 is the permittivity of free space and α is a correction factor, which depends on the dielectric constant of the toner particle. (Hays quotes $\alpha = 1.9$ for a toner particle with dielectric constant of 4. For simplicity we will assume $\alpha = 1$ in this paper). The last column is the ratio of measured to calculated adhesion. It is clear that in all cases the average toner adhesion measured is 7–47 times larger than expected based on the image force F_i calculation, Eq. (1). Hays argues that the observed de-

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◆ IS&T Fellow

* schein@prodigy.com, Independent Consultant, 7026 Calcaterra Dr., San Jose, CA 95120

† Torrey Pines Research, 6359 Paseo Del Lago, Carlsbad, CA 92009
Current address: IBM Corporation, 5600 Cottle Rd., Building 13, San Jose, CA 95193

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TABLE I. Centrifuge Measurements (from Ref. 2)

Size (μm)	Q/M ($\mu\text{C/g}$)	Q/D ($\text{fC}/\mu\text{m}$)	Adhesion (nN), Meas.	Adhesion (nN), Calc.	Ratio, Meas./Calc.
20	5	1	800	17	47
20	10	2	2000	68	29
20	30	6	4000	610	7
10	~0	~0	50	~0	—
10	12	0.62	300	6.6	45
10	5	0.26	43	1.2	36
10	8	0.52	55	4.6	12
10	16	0.84	140	12	12

TABLE II. Electric Field Detachment Measurements (from Ref. 2)

Size (μm)	Q/M ($\mu\text{C/g}$)	Q/D ($\text{fC}/\mu\text{m}$)	Field ($\text{V}/\mu\text{m}$), Meas.	Field ($\text{V}/\mu\text{m}$), Calc.	Ratio, Meas./Calc.
12	4	0.025	5.5	0.26	21
12	9	0.058	9.3	0.61	15
12	13	0.083	13	0.88	15
21	5	0.052	8	0.58	14
9	5	0.023	7	0.25	28

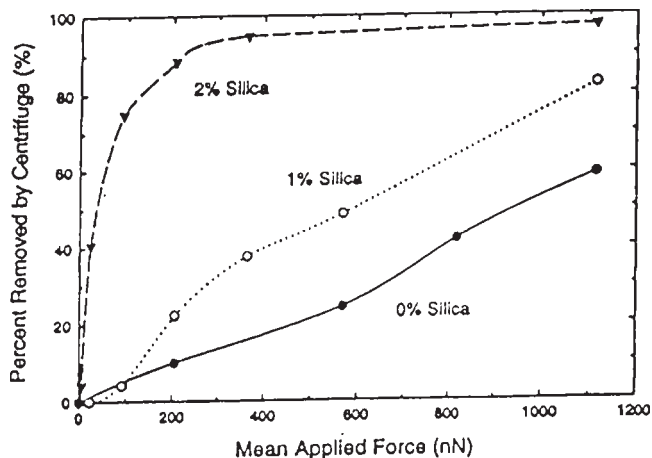


Figure 1. The percent removed by centrifuge as a function of removal force for three levels of silica, 0% (solid circles), 1% (open circles), 2% (solid triangles) (from Ref. 3).

pendence of adhesion force on Q/M , as seen in Table I and II suggests that electrostatic forces are active (despite disagreement in magnitude with the simple model), as opposed to non-electrostatic van der Waals forces.

Gady and coworkers³ published centrifuge measurements showing the full width of the adhesion distribution, although the theory was only applied to the 50% point (Fig. 1). In these measurements silica was added to the toner surface at concentrations of 0, 1 and 2%. These three samples of toner have the same average size (8.5 μm) and the same average charge-to-mass ratio (37 $\mu\text{C/g}$). The expected adhesion image force is 20–40 nN due to image forces, Eq. (1). Clearly much larger values and wide distributions are observed. Clearly silica concentration appears to change the adhesion distribution. They argue that van der Waals forces are needed to explain these results (see below).

Imura⁴ published full adhesion distributions measured by centrifugal measurements (Fig. 2) for a 9 μm diameter toner particle. Again, silica concentration was varied. Again, very wide adhesion distributions are ob-

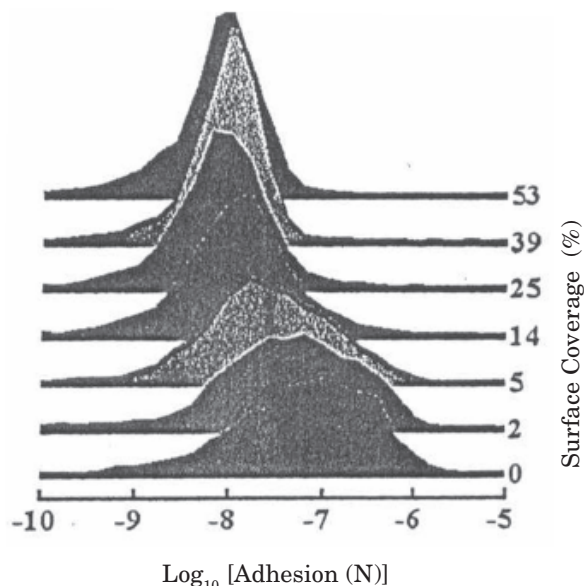


Figure 2. Adhesion distributions for non-tribocharged toners with various toner surface coverage (from Ref. 4)

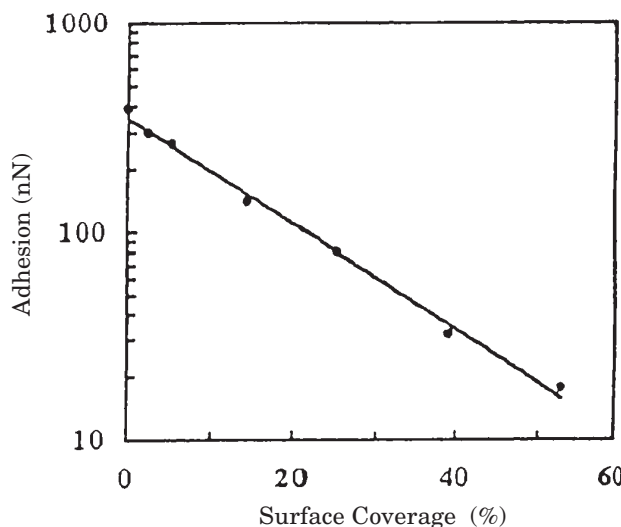


Figure 3. Dependence of the average electrostatic adhesion on the surface coverage for $Q/M = 20 \mu\text{C/g}$ (from Ref. 4)

served, from 0.1 to 1000 nN, 4 orders of magnitude variation. By varying the toner concentration, the average Q/M was varied and it was shown that the average adhesion is proportional to $(Q/M)^2$ consistent with Eq. (1). But the average adhesion also depended on silica coverage at constant Q/M (see Fig. 3), inconsistent with Eq. (1). Equation 1 predicts adhesion of 7 nN for 9 μm particles with Q/M of 20 $\mu\text{C/g}$. The observation (Fig. 3) shows that the average adhesion depends on silica concentration and varies from 17–388 nN. They argue that the dependence of adhesion on Q/M clearly suggests an electrostatic contribution to the adhesion and use Hays' non-uniform charge model.

In a recent article, Hirayama et al⁵ kept the silica surface concentration constant. Again a dependence of adhesion on Q^2 was observed that was about 6 times larger than expected based on Eq. (1).

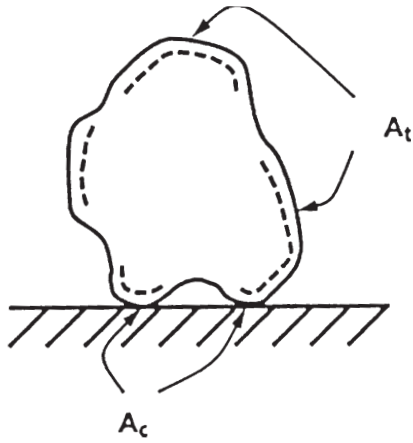


Figure 4. An irregularly shaped toner particle with triboelectrically charged patches of charge density. The total charged area is A , and the sum of the charged areas in contact with the substrate is A_c . (Reproduced from Ref. 2) Copyright 1995; from *Toner Adhesion* by D.A. Hays. Reproduced by permission of Taylor and Francis, Inc., <http://www.rootledge-ny.com>

Measurements of toner adhesion have been reported for ion charged toner.⁶ Electric field detachment experiments provided a measure of the adhesion distribution. The 50% point gave a toner adhesion which agreed with Eq. (1) if the toner charge was 14.2 $\mu\text{C/g}$, which was measured using an electrostatic voltmeter. Subsequent, more direct measurements⁷ gave charge-to-mass ratios of about 2.5 $\mu\text{C/g}$, giving a discrepancy between measured toner adhesion and Eq. (1) similar to results obtained by others.

Published Theories of Toner Adhesion

Non-Uniform Charge Distributions

Hays² non-uniform charge model (charge patch model) starts with the assumption that the toner charge is not uniformly distributed around the surface of a toner particle, but instead is located in patches at the high points (see Fig. 4). He assumes that all of the charge is located in an area which he calls A_t , part of which is in contact with the conductive plane, A_c . The toner adhesion is then assumed to be completely due to the charge in contact with the conductive plane in area A_c , which forms a parallel plate capacitor with its counter charge in the conductive plane. This force (from Ref. 2) is

$$F = \frac{\sigma^2 A_c}{2\epsilon_0} = Qf \frac{\sigma}{2\epsilon_0} \quad (2)$$

where σ is the toner charge per unit area in the charge patch area A_c and $f = A_c/A_t$. Hays then determines that if $\sigma = 1 \text{ mC/m}^2$ and $f = 0.2$, then $F = 100 \text{ nN}$, which is comparable to the measured values in Table I and II.

This result appears to have two problems. Gady and coworkers³ pointed out that the electric field associated with such a high charge density would exceed Paschen breakdown, producing air discharges, discharging the toner the moment it left any surface. Since the electric field in air, E_{air} , is $\epsilon/2\sigma_0$ this is 56 $\text{V}/\mu\text{m}$ for $\sigma = 1 \text{ mC/m}^2$. This value is well above the electric field at which air breaks down at macroscopic distances (3 $\text{V}/\mu\text{m}$).

Second, Schein⁹ has pointed out that if such high electric fields exist on a toner surface, they would exist when the toner was in contact with carrier beads and would therefore affect toner charging behavior. One universally observed toner charge behavior when it is mixed with carrier is the observed dependence of Q/M on toner concentration (the mass of toner on a carrier relative to the mass of a carrier). If such high electric fields existed it would be predicted (see Ref. 9) that Q/M should be independent of toner concentration, inconsistent with the data.

Van der Waals Forces

In order to address the problems with the electrostatic theory of toner adhesion, Gady and co-workers³ proposed that van der Waals forces dominate toner adhesion. The concept is that molecules in close contact across an interface interact by dipole-dipole interactions. These interactions can in principle deform a material around the point of contact. Assuming a deformation occurs, JKR theory can be used to estimate the force of adhesion. It is

$$F = 1.5\omega_A\pi R \quad (3)$$

where R is the effective radius of the asperities of the particle in contact with the plane (toner or silica) and ω_A is the thermodynamic work of adhesion and is related to the surface energies γ

$$\omega_A = \gamma_p + \gamma_s - \gamma_p \quad (4)$$

and their interfacial energy. These values do not appear to vary among materials by more than a factor of four. ω_A was chosen to be 0.05 J/m^2 in Ref. 3. With no silica present, R is the effective radius of the asperities of the toner particle. If silica is present on the toner then R is the effective radius of the asperities of the silica particles (which are much smaller than the radius of the toner asperities) and the number of silica contacts needs to be estimated. The approach used by Gady and co-workers in Ref. 3 is to use JKR theory to estimate the contact radius without silica (196 nm) and then to assume a similar contact region exists when silica is present, allowing an estimate of how many silica particles are in the contact zone. Since there does not appear to be a way for a toner with silica to actually make contact with a plane, this may not be a reliable method of estimating the number of silica contacts and therefore the magnitude of the van der Waals force. Of course, this theory also predicts that toner adhesion is independent of the toner charge, inconsistent with the data shown in Tables I, II, and reported in Refs. 4 and 5, among others.

It appears clear that there is significant disagreement about the source of toner adhesion. While each theory proposed has its strong points, each does not explain some significant experimental result.

Adhesion Theory of a Spherically Symmetric Distribution of Charge Points in Contact with a Conductive Plane

Throughout the toner adhesion papers previously published it always assumed that the adhesion of a spherically symmetric distribution of charge points in contact with a conductive plane is given by Eq. (1). However, it has recently been shown by Czarnecki and Schein⁸ that in contact with a conductive plane, there is an additional force, which has been given the name the prox-

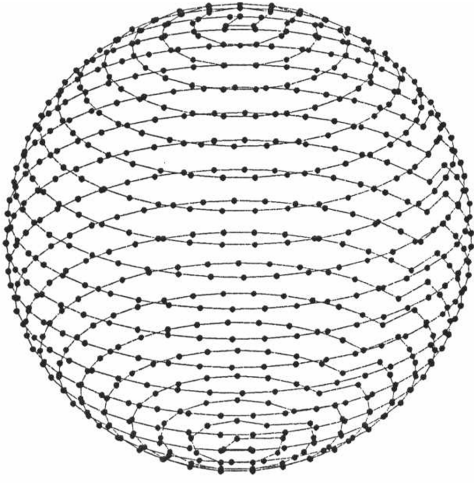


Figure 5. Sphere with charge points (from Ref. 8). Reprinted from *J. Electrostatics*, Vol. 61, with permission from Elsevier, Ltd. Copyright 2004.

imity force (because it is due to the charge in proximity to the contact point) which equals $4/\pi$ times the usual force, Eq. (1).

This result was demonstrated both analytically and with numerical calculations. Finite element analysis was used: the total charge Q was distributed in K charge points uniformly around the surface (see Fig. 5) in annuli parallel to the plane. Then the image charges were located for every charge point in the conductive plane by the standard procedures. Finally the image force was calculated between every pair of charge points. It was found analytically that there were two large contributions to the force. The first was a force between the charge points in the first annuli (the charge points nearest the conductive plane) and their image charges. This is called the proximity force and is equal to

$$F_p = \frac{4}{\pi} \frac{1}{4\pi\epsilon_0} \frac{Q^2}{d^2} \quad (5)$$

Note that Q in this equation is the total charge. Even though the amount of charge in each charge point is only a small fraction of the total charge (Q/K), the large magnitude of the proximity force results from the small amount of charge in each charge point being very close to the conductive plane.

Second, since the charge in the proximity points is much less than the total charge, the rest of the charge can be considered a complete sphere of charge which can be modeled by the usual method, by placing a single charge in the center of the sphere. This gives for the force for the bulk of the charges F_b

$$F_b = \frac{1}{4\pi\epsilon_0} \frac{Q^2}{d^2} \quad (6)$$

The total force F is just the sum of these two forces.

To further verify this result, computer calculations were carried out, again using the charge lumping procedure but this time adding in all of the forces between all of the charge points. In addition the distance from

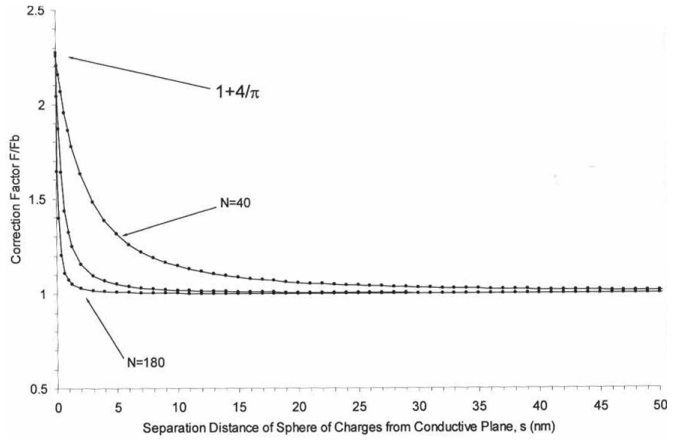


Figure 6. Correction factor to the electrostatic force, normalized to the usual image force (Eq. (6)), versus separation distance s between the sphere and the conductive plane, and N , the number of annuli. The three curves are for $N = 40, 90$ and 180 . (From Ref. 8). Reprinted from *J. Electrostatics*, Vol. 61, with permission from Elsevier Ltd., Copyright 2004.

the sphere to the conductive plane was varied. Also, the number of charge planes (annuli) was varied (from 40, 90, 180) which varied the number of charge points ($2 \times 10^3, 10 \times 10^3, 40 \times 10^3$ approximately). Plotted in Fig. 6 is the correction factor defined as the total force F (Eq. (5) + Eq. (6)) divided by Eq. (6) versus the separation s between the sphere and the plane. As can be seen in Fig. 6, at contact, which is relevant for adhesion theory, the $(1 + 4/\pi)$ result is obtained, verifying the analytical results. But the approach to this value depends on the assumed charge distribution, suggesting that while $(1 + 4/\pi)$ is a universal result for the proximity force when contact is made, its distance dependence depends on the detailed charge distribution. From the point of view of toner adhesion theory, in which the toner particle is in contact with the conductive plane, the new result is that the electrostatic adhesion is the sum of Eqs. (5) and (6). Note that this result does not depend on the assumed charge distribution, i.e., the result is obtained for any N . Taking into account the quantized nature of toner charge, the same result is obtained when the charge in each charge point is one electronic charge. (For the example given in Fig. 6, at $N = 180$ there are 2 charges per charge point for a $12 \mu\text{m}$ diameter toner with charge-to-mass ratio of $12 \mu\text{C/g}$). The electric field at the interface between the toner particle and the conductive plane is now easily calculated: it is just F/Q . For example, for a $12 \mu\text{m}$ particle with $12 \mu\text{C/g}$, the electric field is $0.96 \text{ V}/\mu\text{m}$, smaller than the macroscopic Paschen limit of about $3 \text{ V}/\mu\text{m}$.

This is a remarkable, unexpected result. Only a few charge points with a charge of Q/K which are located in the vicinity of the contact point can generate an attractive force $1.27 (4/\pi)$ times greater than a charge Q located in the center of a sphere of diameter d .

The use of Eqs. (1) or (6) as a model for the adhesion of a charged toner particle to a conductive plane is based on the assumption that a spherically symmetric distribution of charge points can be equivalently replaced with a single point charge in the center of a sphere. It relies

on spherical symmetry to apply Gauss' Law. However, when the spherically symmetric distribution of charge points contacts the conductive plane, spherical symmetry no longer exists and no simple integral can be found to apply Gauss' Law. However, since the conductive plane is an equipotential, the method of images can be used, which is the basis of the approach used here.

While Eqs. (5) and (6) were derived for a conductive plane, it is expected that exactly the same result will be obtained for a dielectric plane (such as a photoconductor) if a multiplicative factor of $(K - 1)/(K + 1)$ is added where K is the dielectric constant of the dielectric. This is because it is well known in electrostatics that image forces are modified by this factor if a dielectric plane is substituted for a conductive plane. Similarly, it is expected that taking into account the dielectric properties of toner will only modify the quantitative results slightly. For example, in Ref. 2 and 10 it is pointed out that $\alpha = 1.9$ for a toner particle with dielectric constant 4. In addition in an electric field detachment experiment the electric field is enhanced due to the dielectric properties of the toner by an amount $\beta = 1.6$. Therefore the net correction factor $\alpha/\beta = 1.19$.

This identification of the proximity force is the basis for a new theory of toner adhesion presented below.

Proximity Theory of Toner Adhesion

Having shown above that a spherically symmetric distribution of charge points has a force of adhesion to a conductive plane that is composed of two parts, one of which is due to a newly identified proximity force, we now apply this result to toner. Assume that toner particles are not perfect spheres and consequently have many contact points. We suggest that at each contact point the proximity force is active. If there are n_p contact points, the electrostatic force of adhesion is then

$$F = \frac{1}{4\pi\epsilon_0} \frac{Q^2}{d^2} + n_p \frac{4}{\pi} \frac{1}{4\pi\epsilon_0} \frac{Q^2}{d^2} \quad (7)$$

At each contact point there is in addition the possibility of van der Waals forces. Therefore the total adhesion force is predicted to be

$$F = \frac{1}{4\pi\epsilon_0} \frac{Q^2}{d^2} + n_p \frac{4}{\pi} \frac{1}{4\pi\epsilon_0} \frac{Q^2}{d^2} + n \frac{3}{2} \omega_A \pi R \quad (8)$$

where n is the number of toner (or silica) asperities which contribute to the adhesion.

It should be clear that the large discrepancy between measurement and calculated adhesion based on the image force of Eq. (1) is easily resolved if n_p is on the order of 10 to 40 contacts, which is not unreasonable. Further,

wide distribution of forces, which are also reported, can be ascribed to large distributions of contact points. Finally, reports in which van der Waals force dominates the toner adhesion can be ascribed to situations in which the R in the last term is due to the asperities on the order of the toner radius. This can occur if the extraparticulates concentration is zero or low enough that toner resin can make contact with the conductive plane.

Summary

A new theory of toner adhesion has been proposed based on the assumption that there is an electrostatic force of adhesion at every contact point, called the proximity force. Because of the many contact points between a toner (or toner with silica on the surface) and a plane it is obvious that this theory can naturally account for the large magnitude of toner adhesion reported in the literature, the wide width of the observed toner adhesion distributions, and the dependence of toner adhesion on Q/M . \blacktriangle

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