Response to Referee 2, handle angeo-2020-23-RC2

"A Comparison of Contact Charging and Impact Ionization in Low Velocity Impacts: Implications for Dust Detection in Space" as submitted by Antonsen et al. to ANGEO

We thank the referee for a number of helpful and interesting comments. In the following, we have tried to respond to all of them, and we have presented the revisions we have carried out in connection to the respective comments. We have not made any major revisions in the overall structure of the paper, only major revisions in the content. We have also revised the abstract.

General comment 1:

We understand that the current presentation of the theory is rather complicated, and may benefit from some revision. Firstly, we strongly agree with the referee that the validity of the model should be discussed in a better manner. We believe that this would also clear up some of the confusion the reader might have. We address this issue first, below, and address the point of a high-level description as brought up by the referee after that.

1. The referee brings up an important point here – the discussion of the validity of the model over the range of parameters used. The intention of utilizing a contact charging model is that it is only really representable for the lower part of the velocity spectrum introduced in the paper (0.1 to 10 km/s). This is not the intention either. What we have failed to convey, based on the confusion brought up by the referee, is however that there *is* a change in charging mechanisms between traditional impact ionization (Langmuir-saha) and capacitive charging. To say however, exactly where the overlap between these two regimes finds place is difficult both experimentally and theoretically. In fact, we do not think there exists an answer in the literature currently that explains the sudden change ("discontinuity") between charge mechanisms at O(1 km/s) impact speeds theoretically, and it is beyond the scope of this work to answer that question.

   Revision: It was intended that the discussion in section 4.1 shall address the validity of an impact ionization vs. contact charging model at 'low' speeds, but we can agree that this discussion is presented too late in the work. We have therefore revised the manuscript by adding a paragraph already in the introduction noting that the current work does not address the exact limit (or speed) that a change between mechanisms happen, only that it happens within the range and that we discuss the arguments for validity of one mechanism before the other [P. 2, L. 19]. Furthermore, we feel that the introduction of section 2 introduces the details of the contact charging—impact ionization dichotomy well.

2. We feel that the essence of Section 2 provides a good high-level introduction into the problem at hand and our solution to it, however, we agree with the referee that the current presentation of the material is not optimal. We feel that a better high-level description/introduction can be given by improving Section 2.

   Revision: At the very beginning of Section 2, we have added what can be considered a high-level 'simple' explanation, which we hope serves the purpose of guiding non-experts towards an understanding of the deeper topics of the paper.

General comment 2:

The referee raises another well founded and important point, which implies that the presentation or rather distinction between the two addressed modes of charging is not sufficient. To clarify, what was intended understood from the manuscript:

> The paper is indeed not comparing apples to apples, as the two discussed mechanisms are fundamentally different. Also addressed in 'comment 1', the two mechanisms in fact compete. To describe the difference at a deeper level than currently, we believe will make the work considerably more complex to comprehend (and probably more speculative). There is no presence of ionization or dissociation at a fundamental level in our contact charging mechanism. Thus, the referee states correctly that the charge production is closely bound to the distribution of fragments. It is also correct that one polarity is completely dominating – which is exactly what has been consistently shown at many instances for rocket experiments and low velocity ice-on-metal collisions in the lab (e.g. referenced works by Havnes, Næsheim, Antonsen et al., Tomsic et al. and so forth). We feel it is also important to point out that the mass spectral characteristica which can be drawn out from conventional impact experiments should not be discussed in connection with our charging model. However, it is merely a tool to better understand when shock wave ionization of certain materials becomes effective/dominant – as we have utilized it in the manuscript.

Revision: In addition to the changes made in connection to general comment 1, which we believe clears some confusion; we have added at the end of the first section of the first paragraph of section 2 [p. 3] an elaboration of the fundamental differences between the discussed mechanisms. This paragraph is subsequently followed by a motivation that we now feel is easier to comprehend and should explain the essence of our modelling efforts better.

General comment 3:

As is hopefully much more clear from the responses above and corresponding revisions in the manuscript, our contact charging model not intrinsically equivalent to ionization as it happens in impacts of small particles with solids. It therefore does not make sense to discuss mass spectral properties of the impact debris of the fragmentation cloud which we provide the model for. There are two important points in the paper where we do in fact touch upon issues related to the role of ions in the impact cloud:

1. In the discussion of the overlap (in produced charge) between impact ionization and contact charging, where it makes sense to note where the literature finds onset of specific ions in impact gas mas spectra. This is interesting, because it is a rough indication of when (shock) impact ionization becomes dominant, and we consequently mention this in section 4.0. We have here relied on work done with the dust accelerator in Heidelberg, and we find the quality and amount of that data to be sufficient for our purpose.

2. In the comparison of our proposed mechanism to the low velocity shock wave ionization solution as presented by Drapatz and Michel. In that regime (i.e. below the limit where mass spectra can detect significant ion partial pressures), particularly volatile ions (e.g. of alkali metals) can diffuse through the molten fragments (or droplets, as Drapatz et al. refers to them) and can be released from their surface. The degree of ionization and produced charge can subsequently be described well with a Saha-Langmuir equation. We address this in depth in section 4.1.

We also want to stress that we do value the tremendous effort behind and results from the Cassini CDA. As the TOF mass spectral properties are not very useful for the direct comparison with our contact charging mechanism, as we have tried to convey in the manuscript, we have not focussed on these. However, from the good advice given by the referee, we understand that a mentioning of such works can easily be justified and have included a reference and description of Hillier et al CDA results in section 4.

General comment 4:

The main issue in this point was addressed in the response to general comment 1, above. We hope it is much clearer as to which regimes of validity we are interested in.

As a side note, the mid-range velocities in which shock wave ionization have been proposed to dominate in (some km/s to a few tens of km/s) have the same velocity-to-charge relationship, or rather power-law, through the whole range, i.e. $Q \sim m^x v^y$. Thus, a somewhat arbitrary upper velocity limit of 10 km/s was chosen. To exaggerate the argument: the limit could have been set to the arguably more arbitrary 9 km/s or 12 km/s – it does not really matter for the presentation of our results. From the discussion in section 4.1, we hope it is clear that we do not propose that contact charging is effective in the upper part of the investigated velocity range. The referee makes a very good point in that the Hertzian deformation theory which we assume cannot be valid at the upper higher speeds investigated here. We have tried to convey this message throughout the paper, but since it may be unclear, we have also emphasised this issue in the presentation of the fragmentation model in section 2.1.

The referee mentions the extreme that at the low end that a grain could behave like a bullet – simply deforming. This is not possible, as the elastic-plastic crossover for grains smaller than several tens of nanometres happens as much lower speeds (even <100 m/s) than interesting for this work [e.g. Rennecke and Weber 2014, Froeschke et al., Tomsic et al.]

Specific comments:

Smaller adjustments; re-phrasing, typos, reusing citations is not addressed below, however, we have tried to follow the recommendations of the referee.

Regarding Saha-solution applied in impact ionization: The referee is correct that a pure Saha-solution is not necessarily valid. What we (and originally Drapatz and Michel) utilize for the low velocity limit of shock wave ionization is the modified Saha-Langmuir equation (now revised in the abstract). This is a

Saha equation that bound electrons in metals are indeed distributed. Such a solution has been shown to hold for impurity ionization in impacts below ~10 km/s, and we present the basics in section 2.3.

Regarding Auer 2012: This is a newer/revised edition (?) It is the preferred citation as listed from Springer Online, at least.

Regarding contact charging: We feel this is now sufficiently answered through the revisions in connection to the general comments above.

Regarding section 2.1: A good point. The citations, as we feel can be understood from the text is used to back up the claims about the degree of fragmentation. Other references in the section help create a more rigorous literature background. An important point is also that key parts of the theoretical background/literature cited in connection to the fragmentation is discussed in section 3.1 (in the Results) where we feel it is much more natural to bring up. *Revision*: This is referenced to in section 2.1, however we have adjusted the first paragraph of section 2.1 to help the reader understand the grounds on which we have chosen modelling parameters.

Regarding Hertzian deformation: revised.

Regarding figure 1: Revised overall description in the text and in the figure caption.

Regarding binning: Misprint. Binning is done with 1 Å spacing, but the sentence was removed as it does is not matter for the end result; a continuum binning (say 0.000…01 nm) would yield very similar results when the fragmenting particles is several nanometres.

Regarding section 2.3: a very good point, brought up by both referees. The reason why we have utilized Mocker et al. results is that the quality of the data is very good in their range of interest. What is not clearly stated here, is that their result is almost “indestinguishable” (for our purpose; Fe-on-Ag) from the findings of Collette et al 2014, who have investigated impacts done to speeds of 2 km/s – which is well embedded into the range of velocities we need comparison data for in order to compare contact charging and impact ionization. Therefore we have used Mocker’s result, however, we could have used Collette et al. with identical conclusions. The data from both Mocker et al and Collette et al for speeds below 10 km/s is furthermore not in the volume ionization regime, which is only expected when impact energies exceed the Fermi-limit (several tens of km/s). Revision: Have tried to specify why we have chosen the Mocker et al results in the start of paragraph 2 of section 3.2.

Regarding Ztot: Bound on fragments. Revised.

Regarding work functions: This is mentioned in the end of the discussion as one of the more uncertain parameters/assumptions, as the work function for nanoscale particles is in fact size dependent to some extent (ref e.g. Wood 81). From mesospheric studies, e.g. Plane et al., it is not necessarily such a large difference for aerosols at sizes of nanometres to tens of nanometres, that it makes a very large difference. Tens of nanometres is by rule of thumb “bulk” as far as cohesive and surface binding energies goes and thus also work function. However, as has also been mentioned, if all modelling parameters are tuned to the extremes of their tolerances, the resulting charge production could change significantly.

Regarding last comment on figure 4: Another very good point that we have had to think about. As far as the contact charging model goes, since it is specific yield (C/kg), it does not matter whether or not the incoming (modelled) projectile is 30 or say 100 nm, since – as shown in the appendix – Qp is proportional to r^3. Thus, the specific yield is the same for any size. A difficult question is then how the plasticity and other modelling parameters change with velocity, which can become complex to give a thought out answer to. We feel it lies a bit outside of the scope of the paper, as the current work is “only” meant to show that we need a better description for the “low velocity” range of impact charging and that our model is a good way to explain some of the observed phenomena connected to such impacts.

Tidbits:

- Charge is not quantized at this point, which may or may not overpredict the contribution from the smaller fragments. Effectively, the charging current can be viewed as a charging probability. A resolution of this ‘issue’ must also discuss in details the role of potential wells on the surface of the smallest fragments – which is probably a phenomenal modelling effort.
- MALDI-type processes are certainly interesting, although may lie a little bit outside our parameter range of interest. In general, the speeds necessary to obtain complete evaporation and have energy available to facilitate MALDI are not obtained at the impact energies we discuss here.