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► **To cite this version:**

Baptiste Chide, Sylvestre Maurice, Agnès Cousin, Bruno Bousquet, David Mimoun, et al.. Recording laser-induced sparks on Mars with the SuperCam microphone. *Spectrochimica Acta Part B: Atomic Spectroscopy*, 2020, 174, pp.106000 -. 10.1016/j.sab.2020.106000 . hal-03492996

HAL Id: hal-03492996

<https://hal.science/hal-03492996>

Submitted on 7 Nov 2022

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Recording Laser-Induced Sparks on Mars with the SuperCam Microphone

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Abstract

The SuperCam instrument suite onboard the Mars 2020 *Perseverance* rover includes a microphone used to complement Laser-Induced Breakdown Spectroscopy investigations of the surface of Mars. The potential of the SuperCam microphone has already been demonstrated for laser ablation under Earth atmosphere in a preliminary study (Chide *et al.*, 2019 [1]) with a small set of samples and fixed experimental conditions. This new experimental study, conducted under Mars atmosphere, explores all the main environmental, instrumental and target dependent parameters that likely govern the laser-induced acoustic signal that will be generated on Mars. As SuperCam will observe targets at various distances from the rover, under an atmospheric pressure that follows diurnal and seasonal cycles, this study proposes a sequence of corrections to apply to Mars data in order to compare acoustic signal from targets sampled under different configurations.

In addition, 17 samples, including pure metals but also rocks and minerals relevant to Mars' surface were tested to study the influence of target properties and laser-matter interactions on the acoustic signal and the ablated volume. A specific behavior is reported for metals and graphite, which rapidly disperse the incoming laser energy through heat diffusion. However, for other minerals and rocks, the growth of the crater is seen to be responsible for the shot-to-shot decrease in acoustic energy. As a consequence, it is confirmed that monitoring the acoustic energy during a burst of laser shots could be used to estimate the

laser-induced cavity volume. Moreover, the amount of matter removed by the laser is all the more important when the target is soft. Hence, the decreasing rate of the acoustic energy is correlated with the target hardness. These complementary information will help to better document SuperCam targets.

Keywords: Mars, Mars 2020 *Perseverance* rover, SuperCam, Microphone, Acoustic, LIBS, Laser ablation, Rock hardness, Penetration depth

1. Introduction

Mars is not as quiet as one can imagine when considering the high sound absorption of the carbon dioxide that composes its low-pressure atmosphere [2]. Indeed, some acoustic waves, especially in the infrasound domain, do propagate and are characteristic of atmospheric phenomena such as dust-devil-like convective vortices [3] or baroclinic waves [4], as detected by the InSight mission. For outreach purposes, infrasounds detected by the InSight Auxiliary Payload Sensor Suite and vibrations captured by the short period seismometer were transposed in the audible. Unfortunately, no real sound recording in the audible range from 20 Hz to 20 kHz has been performed yet.

Scheduled for landing in Jezero crater in February 2021, the Mars2020 *Perseverance* rover will carry two microphones: one to capture acoustic signals during the entry, descent and landing of the vehicle [5] and the other one, part of the SuperCam instrument suite [6, 7], is designed to operate during surface mission, in combination with the SuperCam Laser-Induced Breakdown Spectroscopy (LIBS) technique. This study focuses on the latter which is located on top the rover mast and co-aligned with the LIBS telescope boresight. It will record laser-induced sparks in the 100 Hz to 10 kHz frequency range, during LIBS analysis, with a sampling rate of 25 kHz or 100 kHz. The microphone's primary objective is to support SuperCam's LIBS investigation, but it will also contribute to atmospheric science: it will monitor wind-induced signals to estimate wind speed and direction [8] and it will infer air temperature through the speed of sound when determining the arrival time of the LIBS sound wave [9]. In addition, it will provide diagnosis information on the operation of companion payloads such as MOXIE or the rover's drill.

The intensity of a laser-induced acoustic signal was experimentally shown to be an indicator of the ablation process (see [10] for a detailed review of the experimental applications of shock-waves in laser-induced plasma). More specifically, after firing 10,000 shots on aluminum-oxide ceramics, Grad and Možina [11] noticed a decreasing shot-to-shot evolution of the acoustic signal with different regimes as a function of the number of shots, attributed

29 to different phases of the ablation crater development, whose transitions depend on the tar-
30 get composition. Moreover, the target that had the lowest ablated volume corresponded
31 to the target that had the smallest difference of the acoustic signal amplitude between the
32 first and the last shots. Similar regimes of the acoustic energy along a depth profile have
33 been observed under Mars atmosphere [12], but the link with the ablated volume was missing.

34
35 Our previous experimental study, conducted at Earth pressure and atmospheric compo-
36 sition (Chide et al. [1]), related the laser-induced acoustic signal with laser-induced crater
37 volume and LIBS optical spectrum intensity. For a small set of eight samples, the acoustic
38 energy was shown to decrease with a rate that is dependent on the hardness of the target.
39 For softer targets, the sharp decrease of the acoustic energy was linked with the rapid growth
40 of the laser-induced crater whereas for harder targets, the almost constant acoustic energy
41 corresponded to a low ablation rate. Additionally, a singular behavior of the acoustic en-
42 ergy was observed on the iron-nickel target which had a constant acoustic energy but a low
43 hardness. Correlating the acoustic energy and the ablated volume together has highlighted a
44 linear relationship between these two quantities: by monitoring the relative acoustic energy
45 between the first and the last shots of a laser burst, it is possible to estimate the ablated
46 volume after this given number of shots. These two results represent valuable information
47 in the context of the *in situ* exploration of the surface of Mars with SuperCam for instance
48 to characterize rock coatings [13]. Therefore, it is an expected and necessary step to extend
49 this study to Mars atmosphere (low pressure, CO₂ composition) as the properties of the
50 background medium influence both the laser-induced plasma parameters and the acoustic
51 wave propagation. Moreover, this present work uses a larger set of samples including metallic
52 targets important for understanding the physical mechanisms generating the acoustic signal
53 and with mineral phases expected to be found on Mars in Jezero crater.

54
55 In contrast to laboratory experiments where each parameter can be changed indepen-
56 dently of the others, once on Mars, many instrumental and environmental parameters will

57 change from one target to another. Thus, in order to precisely understand the acoustic signal
58 recorded for several targets of different natures, it is a prerequisite to characterize all the
59 parameters that influence the acoustic signal and estimate their sensitivity. Section 2 details
60 a literature review of these parameters that can be grouped into three categories: instrumen-
61 tal, environmental and target-dependent. After a description, in Section 3, of the setup used
62 in this study, Sections 4 and 5 present the behaviors of the acoustic signal when changing
63 experimental conditions and target nature. Finally, Section 6 summarizes the information
64 on the SuperCam Microphone to support LIBS investigation on Mars, and how the acoustic
65 energy can be used to estimate the target hardness and the ablated volume.

66

67 2. Generation of the Laser-Induced Spark

68 As a laser beam illuminates a sample, its initial energy is converted into heat and trans-
69 ferred within the material. Above a certain threshold energy, ablation of the material occurs:
70 the surface suffers a sudden and sharp increase in temperature leading to a sample mass re-
71 moval due to vaporization. This vapor is ionized and forms a plasma plume whose pressure is
72 significantly higher than the background pressure leading to the formation of a strong shock
73 wave. The whole process is very complex and involves strongly non-linear phenomena. As
74 a matter of fact, laser ablation, vapor expansion and shock-wave propagation are sensitive
75 to parameters from different origins, including: laser beam characteristics that control the
76 energy reaching the sample, target properties that govern the efficiency of the laser-matter in-
77 teraction, and environmental parameters that influence plasma dynamics and the propagation
78 of the acoustic waves. We will discuss these in turn.

79 2.1. Instrumental parameters

80 The laser pulse properties and focusing conditions are key instrumental parameters that
81 control the maximum energy per unit of surface available for target ablation and then strongly
82 influence the LIBS optical and acoustic signal. The ablation process is more efficient when
83 the laser output energy is high. However this process also depends on the surface area
84 over which the energy is spread (giving the laser fluence in J cm^{-2}) and the pulse duration
85 which determines the irradiance (in GW cm^{-2}). For laser pulses lasting a few nanoseconds or
86 longer, the pulse width is also critical, as nanosecond laser pulses partly interact with the ex-
87 panding plasma and one can observe plasma heating and plasma shielding of the surface [14].

88
89 From an acoustic point of view, the sensitivity of acoustic shock-wave parameters with
90 various laser energy and focusing conditions for breakdown of atmospheric air was explored in
91 Manikanta et al. [15, 16]. The peak-to-peak acoustic signal pressure increases linearly within
92 the range of laser energies tested and decreases as the beam diameter at the focal plane
93 increases. Acoustical monitoring of the ablation process [17, 18] showed that there is a linear

94 link between acoustic signal and laser fluence. A similar linear relationship was established
95 between the laser fluence and the ablation rate [19, 20]. The comparison between the four
96 aforementioned studies suggests a potential correlation between the acoustic signal and the
97 ablation rate but no causal link has been clearly established.

98 2.2. Target properties

99 The targeted material properties govern how efficiently the laser power is converted to
100 ablation. These so-called matrix effects tend to complicate the quantitative compositional
101 analysis with LIBS, as the change in laser-matter interactions between targets can affect the
102 amount of ablated mass and the plasma properties [21].

103 Coupling of the energy from the incoming laser pulse depends on the target's optical
104 properties at the laser's wavelength. The laser radiation is absorbed by the material via the
105 Beer-Lambert law over the optical penetration depth δ_{opt} which is defined by the inverse of
106 the absorption coefficient of the sample. The shorter the optical penetration depth is, the
107 more energy per unit of volume is available for ablation. The reflectivity of the material
108 also determines the fraction of the laser beam that is allocated for ablation, however the
109 behavior of this coefficient is sharply changing during the laser pulse due to rapid changes
110 in surface temperature and surface roughening [22, 23]. The laser energy absorbed within
111 the sample is then converted into thermal energy and is propagated into the material via
112 heat conduction depending on its thermal properties. The thermal penetration depth δ_{th}
113 can be defined as a function of the thermal diffusivity and the laser pulse duration: $\delta_{\text{th}} =$
114 $\sqrt{D\tau} = \sqrt{\frac{k}{\rho C_p} \tau}$ with D being the thermal diffusivity, τ the laser pulse duration, k the
115 thermal conductivity, ρ the material density and C_p the heat capacity. For high-diffusivity
116 materials, absorbed heat diffuses quickly, yielding less vaporization [21]. These concepts
117 were used in Fau et al. [24] for the simple case of the ablation of hematite. It should be
118 kept in mind that these aforementioned assumptions apply for purely uniform monocrystalline
119 samples with homogeneous optical and thermal properties. In the case of microcrystalline
120 assemblies likely to be encountered on Mars, laser-matter interaction, laser beam absorption
121 and heat diffusion might be even more complex.

122 Firing successive laser shots at the same location on a target creates a crater that changes
123 the laser-matter interaction. As the laser penetrates into the sample [25], the plasma is more
124 confined inside a cavity, which leads to further changes in its physical parameters [19]. The
125 laser-induced acoustic signal decreases with depth [11], and Chide et al. [1] found a linear
126 relationship between the evolution of the ablated volume and the acoustic signal during a
127 burst of laser shots on the same spot.

128 2.3. Atmospheric conditions

129 The shock-wave expansion has been theoretically described by the Taylor-Sedov blast-
130 wave model [26, 27] assuming that a large amount of energy supporting the explosion is
131 released in an infinitely small volume of perfect gas. This theory was verified experimen-
132 tally [28], showing a hemispherical primary shock front propagating in the ambient gas at
133 supersonic speeds up to $4 \times 10^3 \text{ m s}^{-1}$ with a shock pressure up to 210 MPa. The shock
134 wave becomes increasingly weaker with time and when the pressure behind the shock front
135 approaches the order of magnitude of the atmospheric pressure, the blast-wave model is no
136 longer valid and the pressure perturbation p propagates like a classical spherical acoustic wave
137 [29]:

$$p(r) = \frac{p_0 r_0}{r} \exp(-\alpha r) \exp(i(\omega t - kr)) \quad (1)$$

138 with p_0 the pressure amplitude at a distance r_0 , r the distance from the source, ω the angular
139 frequency, k the wave number and α the total frequency-dependent atmospheric attenuation
140 coefficient. It accounts for energy loss of the wave as it interacts with the ambient medium.
141 Mechanisms involved in the attenuation are absorption due to viscosity, and thermal con-
142 duction, but also rotational and vibrational relaxation of the CO_2 molecules that compose
143 more than 95% of the Martian atmosphere. They also depend on atmospheric pressure and
144 temperature. Semi-empirical models developed by Bass and Chambers [30] and Williams
145 [2] can be used to estimate the pressure damping due to the propagation into the atmo-
146 sphere but have not been validated with experimental data from Mars. Unfortunately the

147 link between the pressure of the shock front close to the source and the associated acous-
148 tic pressure recorded at longer distances is missing. The presence of wind may also affect
149 the recording of laser-induced acoustic waves by producing microphone noise, downgrading
150 the signal-to-noise ratio of the LIBS acoustic detection. However, wind-induced noise has a
151 lower-frequency content that can be filtered out from the raw acoustic signal to retrieve the
152 contribution of laser sparks [12].

153

154 The ambient pressure also plays a significant role in plasma plume confinement [31]. When
155 the pressure is reduced down to 10 Torr, the plume extends to a larger volume, leading to a
156 lower density plasma, optically thin, limiting the plasma shielding. At this pressure, a greater
157 portion of the laser beam reaches the sample surface, allowing more energy for mass ablation.

158

159 To conclude this bibliographic study, all the parameters that were shown to play a role
160 in the acoustic signal are listed in Table 1. It shows how there are coupled together in the
161 different phases that of the acoustic signal generation and propagation.

		Laser Irradiance	Laser-target coupling	Sound Propagation
Instrumental Parameters	Laser Energy	√	-	-
	Focus Quality	√ ¹	-	-
	Distance	√ ¹	-	√ ¹
Material Properties	Optical prop.	√ ²	√ ²	-
	Thermal prop.	-	√ ²	-
	Ablation cavity	-	√ ¹	-
Atmosphere	Composition	-	√ ²	√ ²
	Pressure	-	√ ¹	√ ¹
	Temperature	-	-	√

Table 1: Summary of all the parameters listed in section 2 that are known to play a major role in the laser-induced acoustic signal. They are divided into three categories : instrumental, target-dependent and atmospheric. Each parameter is associated with a phase in the ablation process, from laser energy deposition at the surface of the target to energy diffusion inside the sample and sound generation and propagation. ¹ highlights the parameters whose influence on the acoustic signal is described experimentally by an empirical relationship in the next sections. ² highlights the parameters whose influence on the acoustic signal is assessed qualitatively in the next sections. The other ones were not tested experimentally.

162 3. Experiment description

163 3.1. A Martian LIBS setup combined with acoustics measurements

164 The Mars-atmosphere LIBS calibration test bench at Institut de Recherche en Astro-
165 physique et Planétologie (IRAP, Toulouse, France) used the LIBS capability of the Chem-
166 Cam Mast Unit Engineering and Qualification Model (infrared laser pulse at 1067 nm of
167 about 10 mJ). The Mast Unit was coupled with the ChemCam Body Unit Engineering
168 Model that includes three spectrometers collecting the light emitted from the plasma over
169 the UV (240.1 nm to 342.2 nm), the violet (382.1 nm to 469.3 nm) and the visible plus near
170 infrared (VNIR, 474 nm to 906 nm). More details on this setup can be found in other studies
171 [24, 32, 33]. The full schematics of the experimental bench is represented in Fig. 1. The
172 laser beam exiting the instrument was redirected into a vacuum chamber by a folding mirror
173 which was also used to precisely adjust the pointing of the laser beam onto the sample.

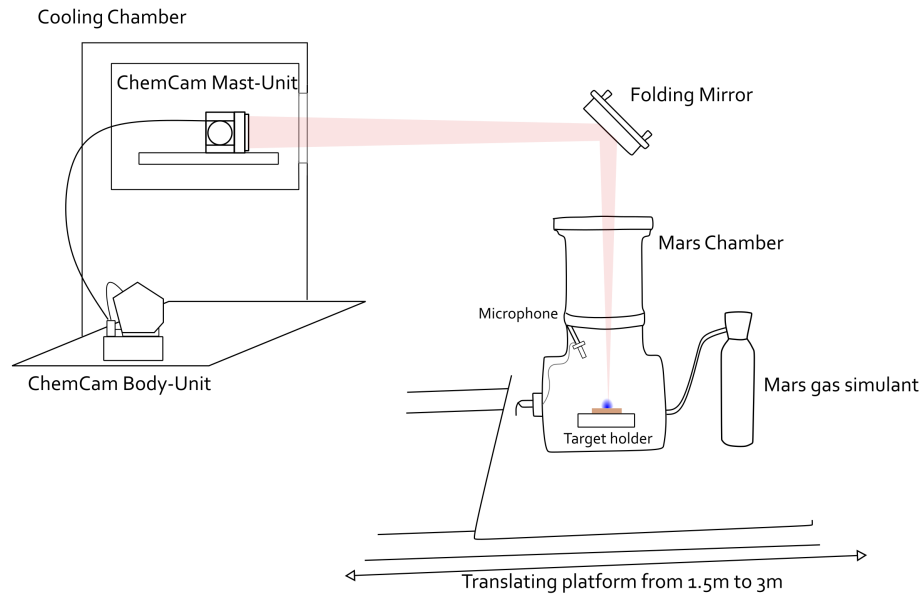


Figure 1: Schematic of the LIBS combined with acoustic test bench under Mars atmosphere.

174 The chamber was filled with a controlled Mars atmosphere (95.7% of CO₂, 2.7% of N₂ and
 175 1.6% of Ar), the pressure of which can be adjusted between 1×10^{-1} and 10 mbar. Targets
 176 were placed on a horizontal aluminum support at the bottom of the chamber and the laser
 177 beam hits the targets perpendicularly to their surface. The chamber was mounted on rails
 178 to move it away from the instrument allowing an extension of the optical path length from
 179 1500 mm to 3000 mm. This facility was also upgraded with a microphone, located about
 180 25 cm away from the target, on the upper part of the chamber. It was pointed toward the
 181 targets. This microphone (same model as the SuperCam microphone) and its acquisition
 182 system were exactly the same ones that were used in [1]. The microphone recorded the LIBS
 183 burst continuously from the first shot to the last one at a sampling rate of 200 kHz.

184 3.2. Set of samples

185 Two different types of samples were used for these experiments. Five pure metals with
 186 tabulated and well known physical properties, and a set of natural homogeneous minerals
 187 graciously obtained from the Collection de Minéraux at Sorbonne Université, Paris, France.

188 They are listed in Table 2, along with basic physical properties that are expected to play a
189 significant role in laser-matter interaction and acoustic signal. References for these proper-
190 ties are listed in the caption of the table. The gypsum, JSC-1 pellet, the black marble and
191 magnetite are exactly the same samples as the ones studied under ambient conditions (here-
192 after defined as Earth atmosphere conditions) [1], and were used as a comparison between
193 the two experiments. The Vickers Hardness was measured with a Micro Vickers Hardness
194 Tester (Buehler MVK H1). The uncertainty in hardness presented in Table 2 is the standard
195 deviation between the 3 to 5 measurements performed on each sample. All the samples were
196 cut or carefully chosen to provide a planar surface to the laser beam and to avoid any surface
197 roughness effects. A picture of all the targets is shown in Fig. 2. Most of the rocks and
198 minerals selected for this study (with the exception of graphite and marbles), correspond to
199 materials that have been identified on Mars [34, 35].

200

201 All the samples were selected for their homogeneity and their variety of thermal and
202 optical properties to be compared with the acoustic signals. In particular, two categories
203 can be formed regarding the thermal and optical penetration depths: metals and graphite
204 that have a long thermal penetration depth compared to the optical penetration depth.
205 These targets easily dissipate laser energy out of the optical absorption zone through thermal
206 diffusion. Other targets have a short heat penetration depth so that the energy deposited by
207 the laser remains localized in the absorption zone [36].

208 3.3. *Experimental procedures*

209 Several experimental protocols have been implemented to test the influence of experi-
210 mental conditions or target properties on the acoustic signal each at a time:

- 211 i The influence of the background CO₂ pressure was tested by increasing the pressure inside
212 the chamber from 1 mbar to 10 mbar by steps of 1 mbar. For each pressure level, a burst
213 of 10 successive laser shots was performed on the titanium target. The location of the
214 impact position was changed between each successive burst to prevent from any cavity

Type	Target	Density (g cm ⁻³)	δ_{th} (nm)	δ_{opt} (nm)	Vickers Hardness	References
Metals	Aluminum	2.70	700	8	340 ± 8	[37]
	Copper	8.96	767	11	88 ± 1	[37]
	Iron	7.87	336	21	104 ± 6	[37]
	Lead	11.35	347	15	8 ± 1	[37]
	Titanium	4.51	216	26	286 ± 38	[37]
Fe-oxides	Hematite	5.15	140	7700	1367 ± 154	[38, 39, 40]
	Ilmenite	4.75	52	8.4 × 10 ⁴	645 ± 76	[38, 39, 41]
	Magnetite*	5.17	92	229	767 ± 134	[38, 39, 40]
Carbon	Graphite	2.16	2497	41	23 ± 3	[37, 42]
Feldspar	Albite	2.62	70	8.5 × 10 ⁶	250 ± 26	[43, 44]
Pyroxene	Enstatite	3.20	91	1.6 × 10 ⁵	49 ± 8	[38, 45]
Sulfate	Gypsum*	1.00	28	-	3 ± 0.5	
Carbonates	Marble	2.71	82	-	124 ± 10	[39, 46]
	Black Marble*	2.69	82	-	177 ± 24	[39, 46]
Rocks	Argilite	2.60	70	-	9 ± 1	[47]
	JSC-1*	1.70	-	-	29 ± 7	
	Basalt	3.00	50	6.6 × 10 ⁴	705 ± 97	[47, 48]

Table 2: Samples used in these combined LIBS and acoustic experiments and some of their physical properties when there have been found. Targets indicated with a star (*) are the same ones that were used in our previous Earth atmosphere study. The JSC-1 target is a pressed pellet of Martian regolith simulant [49] compacted with a load of 3 tons. The gypsum sample is a slice of plaster.

215 effects either on acoustic or spectral intensity. Distance from the instrument to the laser
216 was set to 1650 mm. The acoustic energy and LIBS optical spectrum were monitored for
217 each single shot.

218 ii The influence of the quality of the focus on acoustic data and LIBS spectra can be
219 estimated using a focus stacking (z-stack technique), described in Le Mou  lic et al. [50] for
220 the ChemCam Remote Micro Imager. For the titanium, magnetite and enstatite targets,
221 18 bursts of 10 shots were fired at various distances around the best focus position. The
222 best focus position was determined by the nominal autofocus capability of ChemCam that
223 uses a continuous-wave laser diode. The best focus position is found when the flux back
224 scattered from the target is maximum. This configuration of the telescope maximizes the
225 laser irradiance deposited on the target and therefore it provides the maximum intensity
226 of the emission spectrum [51]. Ten motor steps (corresponding to ~3 mm at this working



Figure 2: Pictures of all the samples detailed in Table 2. The laser craters created for this study are seen in almost all the targets. For some targets, multiple craters resulting from other experiments can also be seen.

227 distance of 1650 mm) separated each consecutive focus distance. The CO₂ pressure was
 228 set to 6.2 mbar for this experiment. Acoustic data and LIBS spectra were recorded for
 229 each laser shot in the same way as previously described. The impact position of the
 230 laser was shifted slightly between each successive burst so that the LIBS craters did not
 231 superimpose each other.

232 iii To test the influence of the laser-to-target distance, the titanium target was set at four
 233 increasing distances from the instrument (1656, 1885, 2402 and 2828 mm). For each
 234 position of the target, an autofocus was performed to measure precisely the optical path
 235 length, followed by 3 bursts of 30 shots. The pressure was set to 6.1 mbar.

236 iv The behavior of the acoustic signal with respect to the nature of the target was tested by
 237 using the full set of samples described in Table 2. Targets were positioned at ~1650 mm
 238 with only slight variations between each distance depending on their thickness (± 10 mm).
 239 For each target bursts of 10, 30, 90 and 150 shots were repeated 2 times (or 3 depending
 240 on the space available on each target) at different locations. The background pressure
 241 was set to 6.2 mbar with variations of ± 0.3 mbar during the time of the experiments.
 242 An autofocus was performed at the center of each target; the precision on the optical
 243 path length is $\pm 0.5\%$ of the total distance [52]. For this experiment, the volume of each

244 cavity was analyzed with a non-contact 3D surface profiler (Sensofar S-NEOX) using
245 either confocal or interferometry scanning. The volume is computed as the integral under
246 the mean plane of the pristine surface. For the lenses used, the uncertainty on the depth
247 of each pixel was $\pm 0.5 \mu\text{m}$. Therefore, the measurement uncertainty on the volume is
248 computed as the surface of the crater multiplied by the uncertainty on the depth. For
249 small craters of $1 \times 10^5 \mu\text{m}^3$ (see craters in metals in section 5.1) the relative uncertainty
250 is about $\pm 50\%$ of the total volume. For craters bigger than $1 \times 10^6 \mu\text{m}^3$ the uncertainty
251 on the volume is lower than $\pm 5\%$. In the following sections, for each figure presenting the
252 ablated volume, a secondary axis displays the associated mean depth which was computed
253 as the average depth between several profiles of each crater.

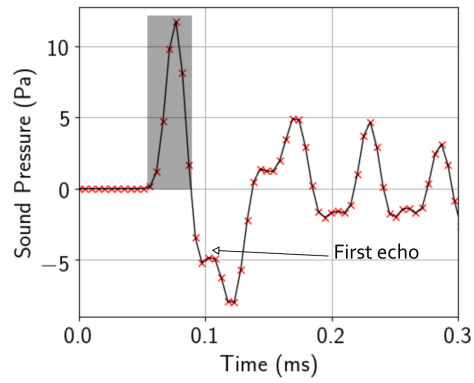
254 3.4. Measurements

255 Unlike our previous study at ambient pressure in which we used an anechoic chamber, such
256 a chamber was not used in this work, as it was not possible to adapt an anechoic chamber
257 for low pressures. The aluminum chamber where the microphone was operated during this
258 study contributed to significant sound reflections. Hence, a careful check of the waveform
259 recorded in the chamber had to be made to be sure that a direct signal was obtained without
260 an echo. Fig. 3a displays a typical LIBS acoustic waveform recorded by the microphone inside
261 the chamber filled with ~ 6 mbar of Mars gas simulant. It shows that the first compression is
262 recorded with no echo superposition and that the first echo arrives $46 \mu\text{s}$ after the arrival of
263 the direct acoustic signal. Considering a sound speed of 273 m s^{-1} at 23°C it corresponds to
264 a propagation distance of 1.2 cm. It matches with a sound reflection on the aluminum plate
265 that holds the samples, the black marble target used for this example being ~ 0.6 cm thick.
266 After that, many echoes are observed in the time series due to multiple reflections on the
267 chamber walls. All the echoes were dissipated before the next laser pulse, which repeated
268 every 333 ms. Two characteristic measurements could be performed on this acoustic wave:
269 the maximum amplitude of the compression (the shock-wave amplitude in Pa) defined as the
270 highest pressure point in the waveform, and also the acoustic energy (in $\text{Pa}^2 \cdot \text{s}$) which is the
271 square value of the waveform, time integrated over the compression phase. This parameter

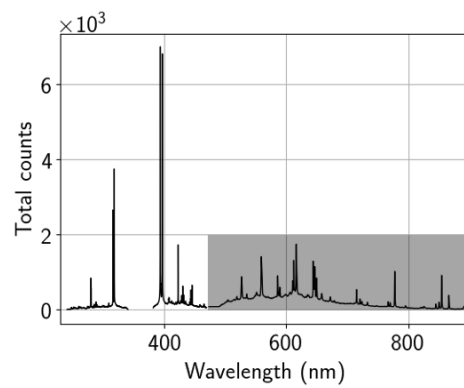
272 was already used in other LIBS acoustic studies [53, 54, 55] over the entire waveform, but
273 here it is restricted only to the compression phase not to integrate echoes. Experimental
274 data show that the acoustic energy is proportional to the square of the shock-wave ampli-
275 tude. Later in this study, the acoustic energy will be presented as a representative parameter
276 of the acoustic signal, as it is computed over more data points than the shock-wave ampli-
277 tude. Nonetheless, each figure will also present the corresponding shock-wave amplitude in
278 a secondary y-axis.

279

280 A typical LIBS optical spectrum acquired by the ChemCam spectrometers is displayed in
281 Fig. 3b for the same shot and the same target as its associated acoustic signal represented
282 in Fig. 3a. Each spectrum is processed following the data pipeline described in Wiens et al.
283 [56], including de-noising and wavelength calibration. However the continuum produced by
284 Bremsstrahlung and recombination radiations was not removed from these spectra. The
285 area under the curve in the VNIR range, including the continuum (spectral region within the
286 shaded rectangle in Fig. 3b) will be used as a parameter representative of the LIBS optical
287 emission intensity because this spectrometer covers the largest spectral range and it has by
288 far the strongest contribution to the continuum [56]. Because the goal of this paper is to
289 study the acoustic signal, this simple spectral parameter will only be used in comparison with
290 the acoustic energy.



(a)



(b)

Figure 3: (a) Typical LIBS acoustic waveform recorded by the microphone at 25 cm for the black marble target inside the vacuum chamber filled with ~ 6 mbar of Mars gas simulant. The shaded area that covers the first compression is the domain used for the computation of the acoustic energy. (b) Typical LIBS optical spectrum acquired for the same laser shot on the black marble. The area below the spectrum in the VNIR range (shaded rectangle) is used as an indicator of the LIBS spectrum intensity.

291 4. Influence of Experimental Parameters

292 Experimental conditions when using LIBS on Mars are always changing depending on the
293 properties of the selected target, and also on the local climate that controls the daily and
294 seasonal cycles of the atmospheric pressure. On the one hand, the irradiance deposited on
295 the target is the key instrumental parameter that governs the efficiency of the ablation. For
296 both SuperCam and ChemCam, the irradiance depends on the offset between the distance
297 retrieved by the autofocus algorithm and the real optical path length between the laser and
298 the sample surface (the quality of the focus for a given distance). But it also depends on
299 the distance from the laser to the target, as the optimal beam radius increases with the
300 distance of the target [52]. On the other hand, the background pressure plays a role in the
301 laser-induced plasma expansion. As these experimental parameters influence the ablation
302 process, they also have an impact on the shock-wave generation and propagation. Therefore,
303 this section studies the sensitivity of the laser-induced acoustic signal with respect to these
304 parameters. Finally, a comparison between the results obtained under Earth atmosphere
305 and Mars atmosphere is presented: these result from changes in both the pressure and the
306 atmospheric composition.

307 4.1. *The impact of atmospheric pressure variation*

308 The variation of the laser-induced shock-wave energy as a function of the background CO₂
309 pressure is represented in Fig. 4 and compared with the evolution of the LIBS optical spectrum
310 intensity. It shows that the acoustic signal is an increasing function of the background
311 pressure, likely due to an increase of the atmospheric density leading to stronger shock-
312 waves. It is obviously silent when the laser is fired under vacuum whereas a plasma is created
313 and its light collected by the telescope. Then the acoustic energy increases linearly between
314 2 mbar and 8 mbar, a range that covers the typical daily and seasonal variations of the Mars
315 atmospheric pressure [57]. It has to be noticed that the acoustic energy increases by ~ 50%
316 between 6 and 8 mbar. The LIBS spectrum intensity also increases linearly but less sharply
317 than the acoustic energy. Above 9 mbar the acoustic energy has lower values than the fitted

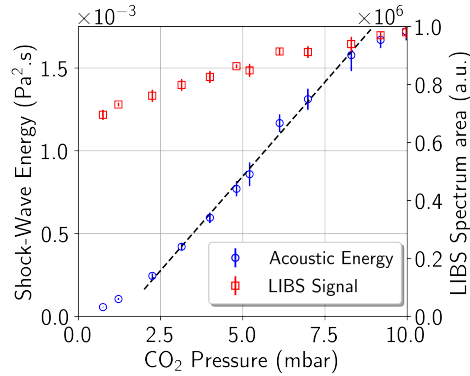


Figure 4: Median acoustic energies over 10 shots at various CO₂ pressure for the titanium target (blue circles). Black dashed line represents a fit of the linear portion of the curve between 2 mbar and 8 mbar ($y = 2.272 \times 10^{-4}x - 2.974 \times 10^{-4}$). It is compared with the median over 10 shots of the LIBS spectrum area (in the VNIR) for each series of shots (red squares). Both for the acoustic energy and the LIBS spectrum area, error bars represent the standard deviation over the 10 shots performed at each pressure.

318 linear model. This pressure may correspond to an increase in collision excitation in the plasma
 319 and the effect of its confinement.

320 4.2. Effects of the quality of the focalisation

321 The LIBS sound level is often used by LIBS teams in their laboratory to rapidly find the
 322 optimal focus of their LIBS setup. In this section the influence of the focus quality on the
 323 laser-induced acoustic signal is investigated. As observed under ambient pressure [58], dif-
 324 ferent acoustic behaviors were observed between irons oxides and other minerals. Therefore,
 325 this experiment was conducted over three targets of different natures: one metal, one iron
 326 oxide and one high absorption mineral (an enstatite).

327

328 Fig. 5 shows the evolution of the acoustic energy for different distances around the best
 329 focus position for the enstatite, magnetite and titanium. Values are normalized by the acous-
 330 tic energy measured at best focus. First, for the three targets, the median acoustic energy
 331 over the 10 shots is at its maximum around the best focus distance and decreases as we move
 332 away from this position. It is observed that out-of-focus laser footprints cover a larger surface.
 333 The laser energy deposited per unit of surface is smaller, leading to a less efficient ablation.

334 It can be noticed that these curves are not symmetric on both sides of the maximum, *i.e.* the
335 mean acoustic energy decreases faster when the focal point is outside the sample than when
336 it is inside. For the titanium target, when the focal point is 10 mm inside the sample, the
337 acoustic energy falls by 17% compared to the one recorded around the best focus, whereas it
338 falls by 50% when the laser is focused 10 mm above the surface of the sample. At best focus
339 a difference in sound level is seen between the three targets: $1.54 \times 10^{-3} \text{ Pa}^2 \text{ s}$ for titanium,
340 $1.25 \times 10^{-3} \text{ Pa}^2 \text{ s}$ for magnetite and $0.44 \times 10^{-3} \text{ Pa}^2 \text{ s}$ for enstatite. It will be discussed in
341 section 6.3.

342

343 The Z-stack analysis is performed in order to measure the LIBS sound's depth of field
344 with respect to the distance from the best focus position. Therefore, the acoustic depth
345 of field is defined as the distance range over which the acoustic energy intensity is above
346 50% of its maximal value. Under these conditions, the values are 45.5 mm for magnetite,
347 33.5 mm for titanium and 14.3 mm for enstatite. At our working distance, it corresponds
348 to an acoustic depth of field of 2.7%, 2.0% and 0.9% of the target distance. This large
349 difference between the enstatite and the two other targets could be explained by a lower laser
350 coupling for enstatite (it has by far the largest optical penetration depth). Therefore, for the
351 enstatite, the irradiance may fall below the ablation threshold faster than for the two other
352 targets.

353

354 Because the acoustic depth of field is a new parameter, it can be compared with the depth
355 of field computed from the LIBS signal that is more often used. Fig. 5 compares the median
356 LIBS spectrum intensity in the VNIR range over the 10 shots with the median acoustic energy
357 as a function of the distance from best focus for titanium, magnetite and enstatite. The LIBS
358 signal follows the same variations as the acoustic energy. For the magnetite, the acoustic
359 energy curve is slightly larger than the LIBS signal intensity curve whereas for titanium the
360 acoustic energy curve is narrower than the LIBS signal intensity curve. However, acoustic
361 and optical spectrum depths of field are very close to each other.

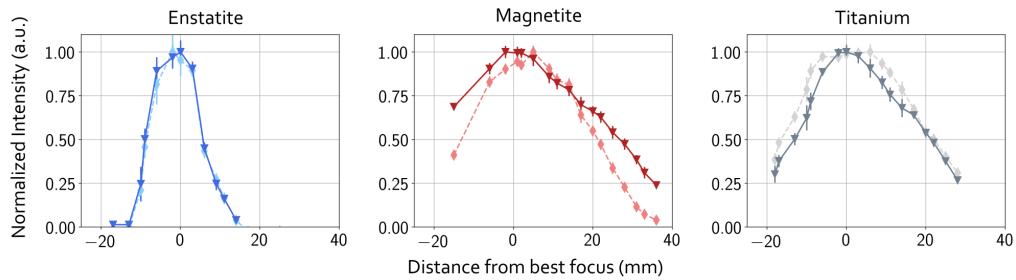


Figure 5: Normalized sound energy (solid darker line) compared with normalized LIBS spectrum intensity (lighter dashed line) at various distances from the best focus position for the three target where z-stacks were performed. Absolute values of the acoustic energy are detailed in the text. The error bars represent the standard deviation of the measurements over the burst of 10 shots. Negative distances correspond to a focal point above the target surface whereas positive distances correspond to a focal located inside the target.

362

363 Consequently, acoustic Z-stacks can be used as a focus method for the telescope and it is
 364 shown to be as accurate as the LIBS spectra Z-stack method that was used with ChemCam
 365 on Mars after the autofocus anomaly occurred (season 2 data, [59]). Acoustic depth-of-field
 366 differences between metallic targets and enstatite, will be considered in Section 6.3 to esti-
 367 mate the uncertainty on the amplitude of the acoustic energy relative to the uncertainty on
 368 the focus.

369

370 4.3. Effects of the variation of the laser-to-target distance

371 Both the focus quality and the laser-to-target distance constrain the irradiance deposited
 372 on the target but the increase of the laser-to-target distance induces an additional effect on
 373 the acoustic signal due to the propagation of sound through a longer path. The impact of
 374 the loss of irradiance with the increase of laser-to-target distance on the acoustic energy and
 375 the LIBS optical spectrum is represented in Fig. 6 for the titanium target. For each distance,
 376 two experimental points are the median over two bursts of 30 shots and two quantities are
 377 represented: the acoustic energy (blue) and the total intensity of LIBS spectra in the VNIR
 378 range (red). It is compared with an estimation of the evolution of the irradiance deposited on
 379 target as a function of the distance for the LIBS setup used in this study, the data for which
 380 are extracted from Rapin [60]. Fig. 6 shows that the acoustic energy decreases the same

381 way as the irradiance whereas the LIBS optical spectrum intensity falls faster. Indeed, the
382 field of view of the collection area of the telescope increases with the distance [52], whereas
383 the plasma size remains the same. Consequently, photon flux collected by the instrument
384 decreases with the distance. This effect is combined with the decrease of the irradiance with
385 the distance.

386 It should be noticed that for these experimental conditions, the distance from the micro-
387 phone to the target is fixed (25 cm), therefore all the points are attenuated the same way
388 by the atmospheric propagation. For this short propagation distance, it can be considered
389 that the atmospheric attenuation is negligible, therefore the acoustic energy is considered
390 not attenuated. However, on Mars, the increase of the instrument-to-target distance will
391 also increase the propagation distance of the acoustic wave, therefore reducing the measured
392 acoustic energy following Equation 1. The blue dashed curve in Fig. 6 represents the acoustic
393 energy, taking into account the propagation distance (the attenuation coefficient considered
394 is 0.05 m^{-1} , extracted from Bass and Chambers [30] at 220 K and 2 kHz). The propagated
395 acoustic energy represented in Fig. 6 is amplified by a factor of 65 for display purposes,
396 to show it on the same plot as the other properties. To give an order of magnitude of the
397 impact of the propagation on the amplitude of the acoustic energy, at 1500 mm the acoustic
398 energy goes from $1.4 \times 10^{-3} \text{ Pa}^2 \text{ s}$ without negligible propagation to $2.24 \times 10^{-5} \text{ Pa}^2 \text{ s}$ when
399 the propagation is taken into account. Therefore, one can note that on Mars, due to sound
400 atmospheric attenuation, the acoustic energy is expected to decrease much faster than the
401 intensity of the LIBS spectrum with the distance from the instrument.

402

403 4.4. Comparison between Earth and Mars atmosphere

404 The difference between ambient atmosphere and Mars atmosphere conditions is provided
405 by the comparison of laser-induced sound evolution for the gypsum, JSC-1, black marble, and
406 magnetite targets that were tested with both setups (see section 3.2). All the instrumental
407 parameters were the same between the two studies with the exception of the optical path
408 length that was $\sim 100 \text{ mm}$ longer for the Earth study. Data under Earth atmosphere are

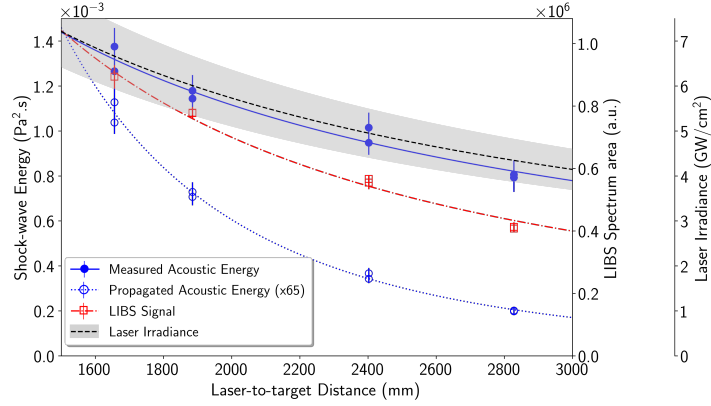


Figure 6: Evolution of the acoustic energy from a plasma on the titanium target as a function of the laser-to-target distance. The microphone was located at a fixed distance of ~ 25 cm from the target inside the Martian chamber. Each experimental point is the median acoustic energy over 30 successive shots at the same location. Both for the energy acoustic and the LIBS signal, error bars represent the standard deviation over the 30 shots performed. Two series of 30 shots were performed for each distance. Experimental points are represented with a power law ax^b that best fits the data (blue solid line, $y = 0.933x^{-0.8853}$). The blue dashed curve is the same acoustic energy but reduced to simulate the propagation into a Mars atmosphere along the distance in the x-axis. In this plot it is amplified by a factor of 65 to compare its evolution with other represented values. The sound attenuation coefficient considered values 0.05 m^{-1} , which is extracted from [30] at 220 K and 2 kHz (see Equation 1 for the propagation law). LIBS spectrum median intensity (in the VNIR range) for each series of shots is represented by red squares also fitted with a power law (dashed red line, $y = ax^{-1.38}$). These experimental results are compared with an estimation of the irradiance on target for this LIBS setup, the data for which are extracted from [60]. The uncertainty on this law (shaded area) is $\pm 11\%$ and is dominated by the uncertainty of the energy of the pulse for this setup.

409 extracted from Chide et al. [1].

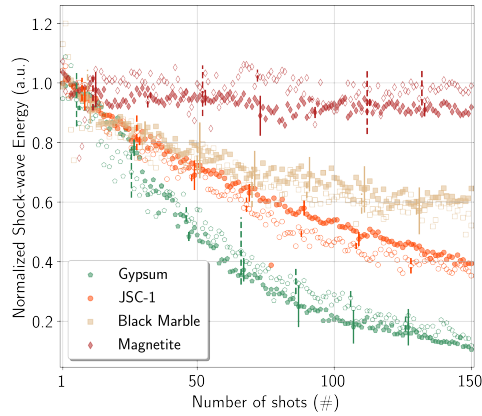
410

411 Figure 7a presents the evolution of the normalized acoustic energy over 150 shots for
 412 the four targets under Mars atmosphere and compared with results obtained under Earth
 413 atmosphere. Although absolute energies are not presented in this figure, the acoustic energy
 414 is about 3 to 9 times greater (depending on the target) under 1 bar of air than under 6 mbar
 415 of CO_2 because of the difference in air density between the two experiments. But more
 416 surprisingly, for a given target, the evolution of the acoustic energy normalized by the first
 417 shot value, follows the same decrease for the two experiments. Looking at the evolution of the
 418 ablated volume could help to understand this similar trend seen between the two atmospheric
 419 conditions. Figure 7b shows the laser crater volume as a function of the number of shots for
 420 the terrestrial and Mars studies. Both experiments show a higher ablation rate during the first

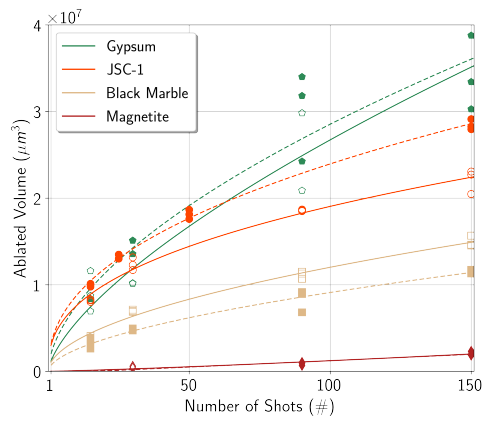
421 ~ 30 shots, then an almost linear increase of the ablated volume for a higher number of shots.
422 More importantly, for each target, the ablated volume is of the same order of magnitude for
423 ablation under a 6 mbar of Mars atmosphere and under 1 bar of air. After 150 shots, the
424 ablated volume in the JSC-1 pellet is 28% larger under Earth atmosphere whereas it is 24%
425 smaller under Earth atmosphere for the black marble compared to the ablation under Mars
426 atmosphere. One could argue that for a higher surrounding pressure, the plasma shielding is
427 more important, leading to a small fraction of the laser beam reaching the surface. However,
428 lida [61] showed that not only does the static pressure play a role in the ablation process, but
429 properties of the surrounding gas such as the thermal conductivity and ionization energy also
430 play a role. For instance the ablation rate under 1 mbar of air is almost the same as under
431 1 bar of helium, with a ratio that also depends on the nature of the target [14]. Therefore
432 the different compositions of air and CO₂, may be responsible for the similar ablation rates
433 observed between the two experiments, counteracting the effects of pressure. In addition, the
434 comparable ablated volumes between Mars and Earth atmosphere explains the same decrease
435 of the acoustic energy as a function of the number of shots. Indeed, it was previously shown
436 [1] that the decrease of the acoustic energy is linearly linked with the ablated volume.

437 For the gypsum target, the deviation between volume measurements for the same number
438 of shots increases significantly after the 90th shot, compared to the other targets. Fig. 8
439 compares typical profiles of laser craters formed on the gypsum and on the JSC-1 pellet,
440 under Mars atmosphere. It shows that after 90 shots, the gypsum crater has a more irregular
441 shape and a larger diameter than the crater resulting from 30 shots. Moreover, the profile
442 has a triangular shape compared to the smooth Gaussian profile for craters created in the
443 JSC-1. It may be due to the brittleness of the gypsum target, the material of which is not
444 only vaporized but also easily ejected with the pressure wave, resulting in a less repeatable
445 volume between two craters with the same number of shots.

446



(a)



(b)

Figure 7: (a) Evolution of the acoustic energy for 150 successive shots at the same location for four targets ablated under Mars atmosphere (filled markers) and Earth atmosphere (unfilled markers). For each target, values are the mean between two (or three) bursts of 150 shots and data are normalized by the energy of the first shot. Error bars, displayed every 20 shots, represent the standard deviation between the three (or two for the magnetite) acquisitions per sample. (b) Evolution of the ablated volume as a function of the number of shots for the four targets compared between Mars (filled markers) and Earth atmosphere (unfilled markers). They are best fitted with a power law ax^b (solid line for Mars atmosphere and dashed line for Earth atmosphere). Note that for the JSC-1 pellet under Earth atmosphere, there was no experiment that created a crater for 90 shots but craters were made of 50 shots instead. For 150 shots under Mars atmosphere, volume measurements for the gypsum are missing. For the magnetite both curves are almost superimpose at this scale.

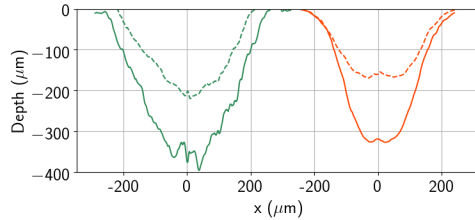


Figure 8: Typical profiles for laser craters made on the gypsum target (left, green) and on the JSC-1 pellet (right, orange) resulting from 30 shots (dashed lines) and 90 shots (solid lines) under Mars atmosphere.

447 5. Influence of target properties

448 This section compares the recorded acoustic energy and the measured ablated volume for
 449 all the targets presented in Table 2, which were sampled under a simulated Mars atmosphere.

450 5.1. The case of metals

451 Here we will discuss metallic targets separately from rocks and minerals, as they are
 452 observed to behave differently and are less relevant to targets expected on Mars, with the
 453 exception of iron meteorites [62] and titanium LIBS calibration targets [63]. Indeed, for
 454 nanosecond laser pulses, the thermal penetration depth is larger than the optical penetration
 455 depth (see Table 2) leading to a greater energy loss into the sample by thermal diffusion [64],
 456 and resulting in melting of some material around the laser-induced crater.

457 Fig. 9 shows the evolution of the acoustic energy and the ablated volume for the five
 458 metals tested in this study. For all the targets, the acoustic energy (Fig. 9a) shows a sharp
 459 decrease during the 3 first shots, and then remains almost constant during the rest of the
 460 burst. Only the lead signal decreases with a linear trend after the initial collapse and the
 461 copper signal slightly increases after 70 shots (see arrow highlighting this trend in Fig. 9a).

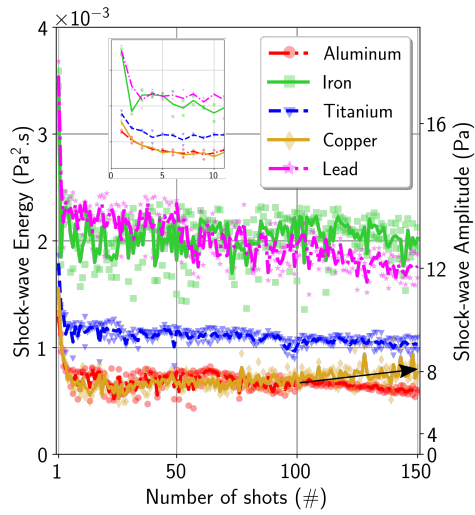
462 Craters formed in the lead sample are deep, with a conical shape and they display a
 463 rim which is higher than the pristine surface. Craters on other metals are shallow, and at
 464 their bottom, they present an irregular floor made of molten-like metal. Some molten ejecta
 465 material is seen near the location of the impact as if it was ejected during the pulse. Moreover,
 466 the ablated volume of copper, iron, titanium and aluminum is very small: the maximum is

467 $2 \times 10^6 \mu\text{m}^3$ for 150 shots on aluminum compared to more than $1 \times 10^7 \mu\text{m}^3$ for the same
468 number of shots for lead. This large amount of vaporized lead compared to other metals
469 was also observed in Iida [61] and attributed to lower change-of-state temperatures for lead
470 compared to other metals.

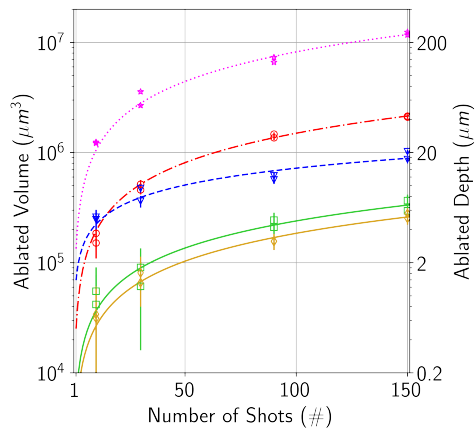
471 Considering these observations, the shallow ablated craters explain that the acoustic
472 energy is almost constant for iron, copper, aluminum and titanium, contrary to lead where
473 its linear decrease could be explained by the higher ablation of the sample that created a
474 deep crater. The large drop in acoustic energy during the 3 first shots, characteristic of
475 metals, was already observed on aluminum by Lu et al. [65], and was considered as surface
476 cleaning, such as the removal of a thin oxide layer. The same study also noticed the constant
477 regime of the acoustic energy for higher numbers of pulses. In a follow-up study focused on
478 copper samples, Lu et al. [53] noticed the increase of acoustic energy specific to copper and
479 attributed it to surface morphology changes inside the laser spot, leading to modification
480 of copper optical properties. Indeed, pristine copper has a very high reflectivity coefficient
481 compared to other metals tested here ($R=0.94$ at 1067 nm) and it must drop to significantly
482 lower values with the roughening of the surface due to ablation. Therefore, for copper, the
483 reduction of the reflectivity coefficient increases the fraction of laser energy available for
484 ablation. The difference in absolute values of the acoustic energy between different metals
485 shown in Fig. 9a was also pointed out in Lu et al. [65] but was left unresolved. It will be
486 discussed in Section 6.3.

487 5.2. The case of minerals and rocks

488 Depth profiles of 150 shots were conducted on all the other targets; the acoustic energy
489 as a function of the number of shots is represented in Fig. 10 only for five of the targets. The
490 absolute acoustic energy of the beginning of the sequence differs depending on the nature
491 of the target. Most of the targets are grouped between $1 \times 10^{-3} \text{Pa}^2 \text{s}$ and $2 \times 10^{-3} \text{Pa}^2 \text{s}$
492 (corresponding to amplitudes from 9 Pa to 13 Pa) whereas the marble and the argillite lie
493 around $3.2 \times 10^{-3} \text{Pa}^2 \text{s}$ (equivalent to 17 Pa) and graphite at $7 \times 10^{-3} \text{Pa}^2 \text{s}$ (27 Pa). This
494 variation of the absolute acoustic energy, possibly a function of the nature of the material, is



(a)



(b)

Figure 9: Evolution of the acoustic energy and shock-wave amplitude (a) and the ablated volume and depth (b) over 150 shots for the five metallic targets. For the acoustic energy, the two series of 150 shots at 2 different locations are represented by the symbols and the mean over these two series is represented by the colored line. The inset in (a) shows a close-up view over the first ten shots. The black arrow highlights the increasing trend for copper. For each target, two craters with the same number of shots are performed. The volume is fitted with a power function ax^b . Details on the error bars for the volume measurements are provided in Section 3.3. The logarithmic scale representation makes the error bars of the biggest craters barely visible.

495 discussed in Section 6.3, considering the sensitivity of the acoustic signal with regard to the
496 quality of the focus, the background pressure and the distance.

497

498 Our previous study performed under Earth atmosphere has shown that the evolution of
499 the acoustic energy could be represented by a decreasing exponential function; its exponential
500 decay rate is correlated with the hardness. Here under Mars atmosphere, as with the Earth
501 atmosphere experiments, the decay rate of the acoustic energy is a decreasing function of the
502 hardness. The acoustic energy of graphite (soft mineral) has dropped by 30% in 150 shots
503 whereas it has been reduced by only 6% for hematite (hard mineral). Although a decreasing
504 exponential function was used to represent the evolution of the acoustic energy over 300
505 shots under Earth atmosphere, it does not perfectly fit this new data set, especially during
506 the first shots. Indeed, targets can be grouped into two categories regarding the shot-to-shot
507 evolution of the acoustic energy: the first group, which corresponds to the softer targets,
508 shows a slope change of the acoustic energy after the first tens of shots. This can be seen for
509 instance for the graphite, marble and gypsum in Fig. 10, with slope changes observed around
510 the 20th, 50th and 70th shots, respectively. Targets belonging to this first group are gypsum,
511 JSC-1, graphite, argillite, marble, black marble and basalt. This characteristic evolution of
512 the acoustic energy can be fitted with two linear functions, one before and one after the
513 slope change. For this set of targets, the acoustic energy decreases faster during the first
514 regime than after the slope change. The second group is composed of harder targets where
515 no slope change is seen (hematite, magnetite, ilmenite, enstatite and albite); see for example
516 the magnetite in Fig. 10. It can be fitted with a linear function over the entire 150 shots of
517 the burst.

518 The presence of a rapidly decreasing regime followed by a lesser slope regime had already
519 been noticed by Murdoch et al. [12] for soft targets and in Chide et al. [1], not only with the
520 acoustic energy, but also with LIBS spectral data. It was attributed to the growth of the laser
521 crater leading to a loss of laser-material coupling due to both plasma shielding and steeper
522 crater walls. Therefore, the evolution of the acoustic energy has to be studied in comparison

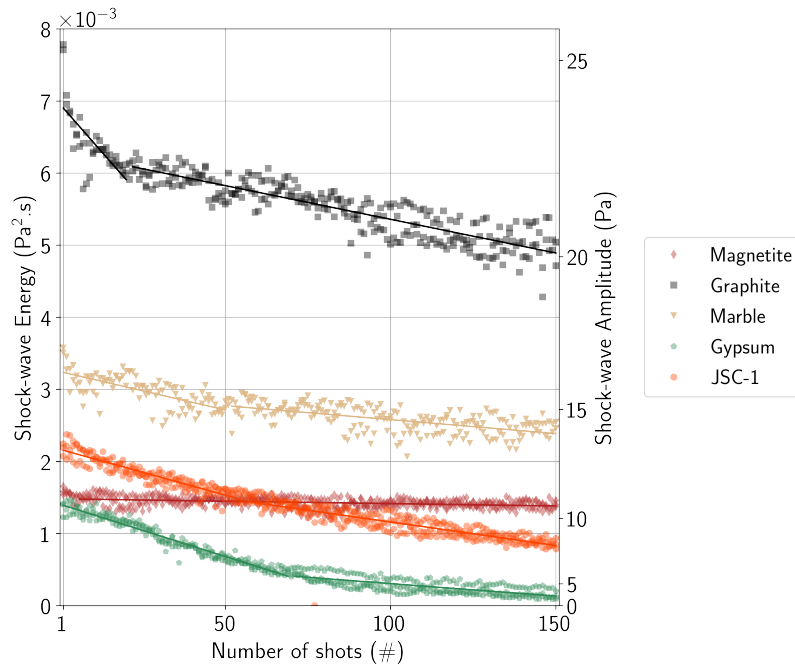


Figure 10: Variation of the acoustic energy (and shock-wave amplitude) during a burst of 150 consecutive shots on several targets. For each target the 2 (or 3) series of 150 shots at different locations are represented. Depending on the target, the mean of these series is fitted with two successive linear functions (softer materials, see examples given for graphite, marble, JSC-1 and gypsum) and with only one linear function for harder targets (see example given for magnetite). Other targets are not represented here but they behave similarly and all have their first shot acoustic energy between $1 \times 10^{-3} \text{ Pa}^2 \text{ s}$ and $2 \times 10^{-3} \text{ Pa}^2 \text{ s}$. Normalized evolution of acoustic energy for targets that are not represented here can be found in Fig. 12 and in Fig. 15 for the first shot acoustic energy for each target. Acoustic energy for metal is not displayed in this plot but is represented in Fig. 9a.

523 with the ablated volume.

524

525 The evolution of the ablated volume as a function of the number of laser shots that built
 526 the crater is given in Fig. 11. The same behavior is observed for all the targets including
 527 the metals: the ablation rate is higher during a first phase lasting about 30 shots. Then
 528 it is followed by an almost constant ablation rate phase, leading to a linear increase of the
 529 volume (see also evolution of the ablated volume in linear scale in Fig. 7b). As for the
 530 evolution of the shot-to-shot ablated volume, two groups of targets can be identified in Fig.

531 11: targets with an ablated volume higher than $4 \times 10^6 \mu\text{m}^3$ after 150 shots (upper part of
532 the figure, starting from the basalt) and a second group with targets with an ablated volume
533 lower than $2 \times 10^6 \mu\text{m}^3$, that includes iron oxides, graphite and metals with the exception
534 of lead. Notice that, due to the representation of the volume in a logarithmic scale, the
535 evolution of the ablated volume for targets in the second group is almost flat compared to
536 targets belonging to the first group.

537 Those two groups of ablated volume almost concur with the two groups of targets high-
538 lighted in Fig. 10 for the acoustic energy. Targets with a high ablated volume show a slope
539 change in the shot-to-shot evolution of the acoustic energy whereas no slope change is seen
540 in acoustic data for targets with a lower ablated volume, with the exception of albite and
541 graphite. For the albite, no slope change was observed for the acoustic energy, but for this
542 target (and also for the enstatite) the normalized standard deviation of the acoustic energy
543 over the 150 shots is at least twice as high as other targets (see normalized acoustic energy
544 for albite and enstatite in Fig. 12). This can be explained by the high optical penetration
545 depth for enstatite and albite (see Table 2) leading to a less efficient laser-to-target coupling.
546 Therefore, the shot-to-shot acoustic energy varies much more than for other targets, leading
547 to a bias in the slope retrieval. For the graphite, a slope change around the 20th shot is
548 noticed on acoustic data whereas the ablation volume is of the order of magnitude of iron
549 oxide target. As was mentioned for the other metals (see section 5.1), this may be explained
550 by the thermal penetration depth for graphite (2497 nm [37]) that is 60 times higher than
551 its optical penetration depth (41 nm [42]). Therefore, the energy absorbed by the graphite
552 is dissipated through heat conduction leading to a lower amount of energy partitioned to
553 ablation. Considering its physical properties, graphite has a behavior comparable to metals.

554
555 For the first group of targets identified in Figs. 10 and 11, the softer ones, the ablation
556 rate is high, especially during the first tens of shots: from $\sim 40 \mu\text{m}$ for basalt up to $\sim 300 \mu\text{m}$
557 for gypsum after only 30 shots. Thus, as the crater is growing quickly, some cavity effects
558 rapidly reduce the laser-material interaction when the number of shots increases, leading to a

559 rapid decrease of the acoustic energy: the plasma is more and more confined, hence denser,
560 enhancing its shielding of the beam. A smaller fraction of the laser energy reaches the target,
561 weakening the shock-wave. In addition to that, and to a lesser extent, the laser no longer
562 impacts a flat surface orthogonal to the beam, but instead is incident on tilted crater walls,
563 leading to a geometric reduction of the irradiance deposited when the laser is fired inside a
564 cavity. After a given depth that may depend on the nature of the target, the plasma shield-
565 ing may have reached a maximum and only the geometric reduction of the laser-material
566 coupling decreases the acoustic energy, possibly explaining the slope change seen on acoustic
567 data. For the second group of targets, the harder ones, the laser crater is shallow (less than
568 $\sim 10\ \mu\text{m}$ after 30 shots) likely leading to an almost constant shot-to-shot plasma shielding.
569 Variations of plasma shielding with the number of shot cannot be seen in acoustic data. Only
570 changes of surface properties and crater formation downgrade the laser-target interaction and
571 are responsible for the slight decrease of the acoustic energy.

572

573 These assumptions regarding the plasma parameters can be compared with the LIBS
574 optical spectrum intensity itself. Fig. 12 compares the shot-to-shot evolution of the acoustic
575 energy with the evolution of the LIBS spectrum intensity in the VNIR range. Both values are
576 normalized to corresponding values for the first shot. Targets are arranged with respect to
577 the ablated volume: the two first rows in Fig. 12 group together targets with a high ablated
578 volume with the addition of the enstatite for which the volume was impossible to measure.
579 The last row puts together targets with a lower ablated volume: iron oxides and graphite.

580 As was already observed for samples targeted under Earth atmosphere, the evolution of
581 the optical spectrum intensity differs relative to that of the acoustic energy depending on the
582 amount of material ablated from each target. For targets which have high ablation rates (the
583 two first rows in Fig. 12), the acoustic energy decreases, whereas the LIBS spectrum intensity
584 increases. This increase may indicate a denser plasma as it is more and more confined as
585 the cavity grows. Moreover, after 150 shots, the difference between the normalized LIBS
586 optical spectrum intensity and the normalized acoustic energy is all the more important as

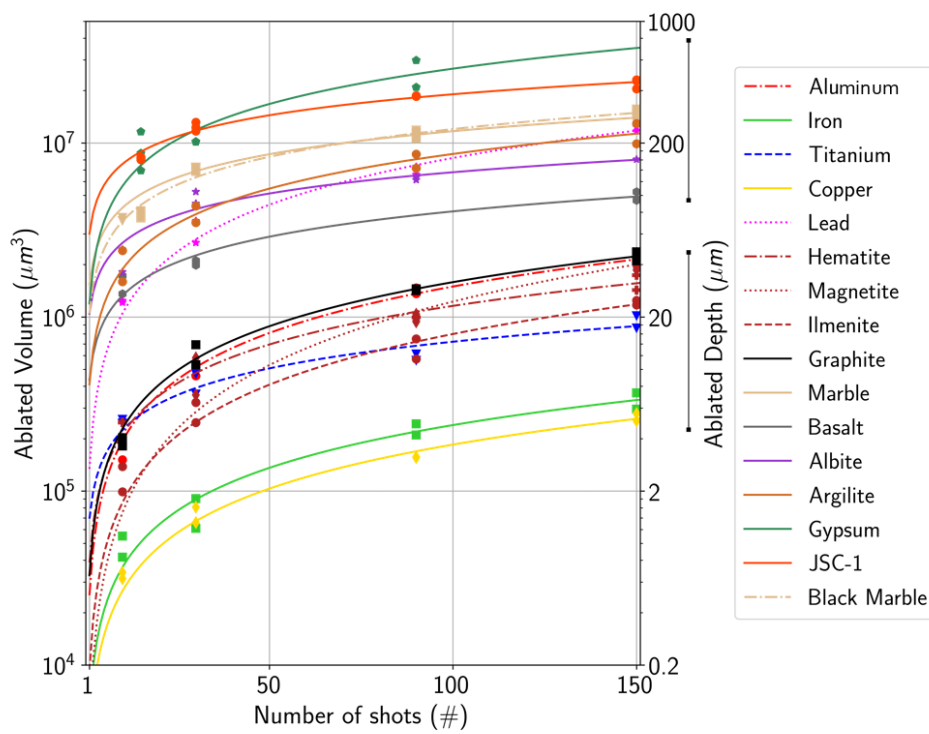


Figure 11: Evolution of the ablated volume (and depth) as a function of the number of shots for 17 of the 18 targets tested (including metals). There are 2 (or 3) repeated points per target for a given number of shots that created the crater. The volume for enstatite is not represented because of poor measurements. Measurement uncertainties are not represented as they are barely visible for volumes higher than $1 \times 10^5 \mu\text{m}^3$. They are displayed for metals in Fig. 9b. Details on measurement uncertainty on the volume are provided in Section 3.3. Points for all other targets are best fitted with a power law ax^b (colored lines). Vertical bars on the right of the figure show the two groups of targets discussed in the text with respect to the ablated volume.

587 the ablated volume is high. For instance, the basalt is the target belonging to this group
588 that has the lowest measured ablated volume. It is also the target from this group that
589 presents the smallest gap between the normalized acoustic energy and the normalized LIBS
590 optical spectrum intensity. For gypsum, the LIBS spectrum intensity starts decreasing after
591 50 shots. This effect, which was also observed under Earth atmosphere, is likely due to the
592 crater shape anomaly highlighted in Fig. 8. After 50 shots the laser may ablate torn walls and
593 some matter that is loosely consolidated or collapsed at the bottom of the crater, resulting
594 in a more rapid loss of coupling for this target. For targets with a low ablation rate (last
595 row in Fig. 12), the LIBS optical spectrum intensity seems to follow the same variation as
596 the acoustic energy. As laser craters formed in these targets are shallow, no cavity-induced
597 effects are seen in the LIBS optical trends.

598 As a conclusion, the evolution of the LIBS spectrum can be explained by changes of
599 plasma properties when it is produced inside a cavity. This explanation is consistent with
600 the hypothesis made above stating that the shot-to-shot decrease of the acoustic energy is
601 a consequence of the growth of the laser-induced cavity. Therefore, the next section applies
602 these findings to the study of the Martian targets to be analyzed with SuperCam on Mars.

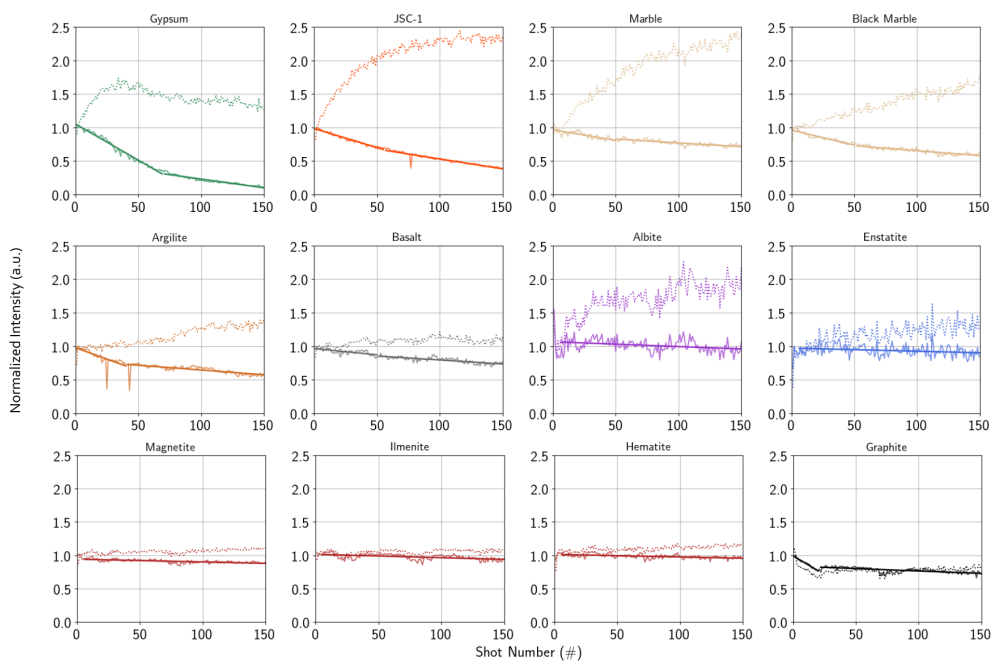


Figure 12: Shot-to-shot evolution of the acoustic energy normalized by the first shot (solid line) compared with the evolution of the normalized LIBS spectrum intensity in the VNIR range, including the continuum in the VNIR range, including the continuum (dotted line). Targets that have a high ablated volume are grouped in the two first rows, with the addition of the enstatite for which the ablated volume measurement was impossible but that presents an increasing continuum. The last row groups targets that are shown to have a lower ablation rate. The portions of linear functions that fit the evolution of the acoustic energy is also represented.

603 **6. The acoustics as a support to LIBS investigations on Mars**

604 The combined study of the acoustic energy and the ablated volume has shown that the
605 decrease of the former is a tracer of the ablated volume: the softer the target, the higher
606 the ablated volume and the faster the decrease of the acoustic energy. Hence, tracking the
607 acoustic energy along a LIBS burst can give information about both the target hardness and
608 the ablated volume.

609 *6.1. Inferring target hardness*

610 It has been observed that the shot-to-shot acoustic energy can be represented by portions
611 of linear functions, two for softer targets and only one for harder materials. Linear functions
612 are expressed with the following expression $E = E_0(1 - m)$ with m given as the normalized
613 linear decay rate. In order to be as close as the usual operational conditions of SuperCam on
614 Mars, for each target, the linear function is fitted only over the 30 first shots (typical number
615 of shots performed on Mars). Only for the graphite whose slope change in the acoustic
616 energy occurs before the 30th shot, the linear function is fitted from the first shot up to the
617 transition (20th shot). This linear decay rate m of the acoustic energy is represented as a
618 function of the Vickers hardness measured for each target in Fig. 13. It is also compared
619 with the same measurements under Earth atmosphere extracted from our previous study for
620 gypsum, the JSC-1 pellet, black marble and magnetite. This figure shows that the decay
621 rate of the acoustic energy is a decreasing function of the Vickers hardness. This coefficient,
622 expressed in shot^{-1} is independent from the absolute amplitude of the acoustic energy but
623 also almost the same for an ablation under Earth atmosphere and Mars atmosphere. Only
624 points for the enstatite (blue square) and the albite (purple star) do not follow the same trend
625 as other targets and have a larger dispersion. Indeed, the relatively high standard deviation
626 of the acoustic energy for these two targets, likely due to high optical penetration depth, may
627 lead to a bias in slope retrieval. For this type of targets, there is a risk of misinterpreting
628 the data that could be evaluated with the dispersion of the acoustic energy along a burst. It
629 may also be difficult to distinguish the harder materials with this method but only infer that

630 they have a Vickers hardness higher than 500.

631

632 However, Fig. 13 confirms that the decrease rate of the acoustic energy can be used to
633 estimate rock hardness at remote distance on Mars. It extends under Mars atmosphere and
634 with a larger set of targets our previous results obtained under Earth atmosphere. Moreover,
635 as the linear decrease of the acoustic energy is extracted from a fit from the first shot of
636 a burst to a slope change in data, if it occurs, it does not require long sequence of shots.
637 Therefore, on Mars with SuperCam, it will be possible to estimate hardness on the majority
638 of targets as during the 8 years of activity on Mars, most of the ChemCam targets were fired
639 with bursts of only 30 shots.

640 It should be noted that other approaches exist to estimate target hardness that are only
641 based on the LIBS spectrum: the ratio of ionic to atomic emission lines [66, 67] but also the
642 plasma temperature [68, 69] were shown to linearly correlate with the hardness of selected
643 targets. This is likely the result of a faster shock-wave for the harder materials that leads to a
644 more ionized plasma [70]. The approach chosen in our study only relies on the acoustic energy.

645

646 Looking at the results with the Mars Science Laboratory rover at Gale crater, each ge-
647 ological formation can cover a large area (km^2) and so a Mars rover may travel for many
648 months in the same formation. However, that formation can have variations in hardness
649 that can affect drilling capabilities and which are also of geological interest. These hardness
650 variations can occur due to changes in pore-filling cements of sediments and/or alteration.
651 Vera Rubin ridge, which formed through deflation of softer surrounding sediments, is one
652 example [71, 72], where the ridge presented difficulties in drilling, and the ridge's physical
653 properties were also of strong scientific interest [73]. In our study, we investigated a group
654 of diverse rock and mineral types. An investigation using various samples that are all from
655 the same sedimentary formation but have different hardness values may show a much tighter
656 correlation than that seen in Fig. 13. Such a study may provide a better estimate of the
657 accuracy of the hardness values that can be determined from SuperCam in a realistic setting.

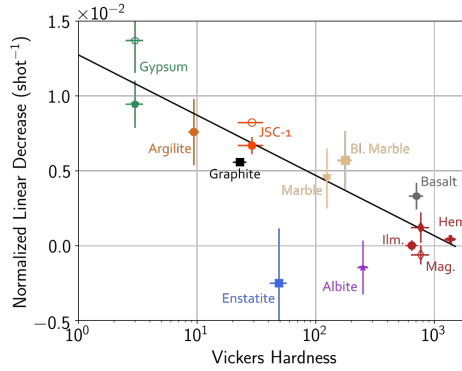


Figure 13: Linear decay rate of the acoustic energy measured for the 12 minerals and rocks tested in this study under Mars atmosphere (filled markers) as a function of the Vickers hardness measured for each material. It is compared with linear decay rate computed for the same gypsum, JSC-1, black marble and magnetite whose data are extracted from our previous study under Earth atmosphere (unfilled markers). Linear decay rate is computed by fitting a linear function over the 30 first shots for bursts of 30, 90 and 150 shots. If the slope change observed on the acoustic energy occurred before the 30th, the fit is performed up to the transition. Vertical error bars represent the standard deviations of the linear decay rates retrieved for the bursts of 30, 90 and 150 shots on each target. Horizontal error bars represent the standard deviation between the 5 hardness measurements performed per target. Marker shape and color follow the same color code than in Fig. 10. Both data sets are fitted with a straight line (solid line for Mars data, dashed line for Earth data). Hem: Hematite, Mag: Magnetite, Ilm: Ilmenite.

6.2. Estimation of the ablated volume

Our previous study under Earth atmosphere has demonstrated that the relative decrease of the acoustic energy was linearly linked with the ablated volume and that this relationship was quite independent of the target properties. For each rock and mineral tested in this study, Fig. 14 shows the acoustic energy of the last shot of the sequence that created a given crater as a function of the measured ablated volume of this crater. Acoustic energies are normalized by the first shot of the sequence. It is compared with results under Earth atmosphere [1], for the four targets that are in common between the two studies. All these data follow a linear trend confirming that the decrease of the acoustic energy can be used to estimate the ablated volume after a given amount of shots. As reported as part of the comparison with the ablation under Earth atmosphere (see section 4.4), both the excavated volumes and the acoustic energies were similar, so it makes sense that both results under Earth and Mars atmosphere follow the same trend. It can be noticed that for the graphite sample, although the acoustic energy is a linear function of the ablated volume, the acous-

672 tic energy decreases faster than for other minerals. This may be explained by the singular
673 thermal behavior of graphite. The laser was fired perpendicular to the graphene planes that
674 have a very high thermal conductivity [37]. Therefore heat must have been dissipated by
675 the sides of the crater. For volumes lower than $1 \times 10^7 \mu\text{m}^3$ the dispersion around the fit-
676 ted line is larger than for higher volumes. It may be due to surface roughness that affects
677 volume measurements in a larger extent for shallow craters than for deep ones. With Su-
678 perCam on Mars, most of the targets will be ablated with burst of 30 shots but with a
679 laser energy greater than 24 mJ [6] that might result in deeper craters: laser-induced craters
680 created with 30 shots on a basalt sample with the qualification model of SuperCam were
681 $120 \mu\text{m}$ deep [7] compared to $\sim 40 \mu\text{m}$ with the laser used in this study. However, bursts of
682 150 shots or more will be possible with SuperCam to create deeper craters on specific targets.

683

684 Nevertheless, this linear relationship between the ablated volume and the acoustic energy,
685 observed under Earth atmosphere, is confirmed and extended under Mars atmosphere for
686 most of the rocks and minerals tested in this study. It demonstrates that the SuperCam
687 microphone has the ability to estimate the ablated volume of LIBS targets, and consequently
688 the crater depth. This is valuable information in order to study the chemical stratification
689 with depth, especially to constrain the thickness of rock coatings [74]. The volume estimation
690 would also be critical to discuss the heterogeneity of a target that presents variations in its
691 shot-to-shot LIBS signal.

692 *6.3. Sensitivity of the measurements expected on Mars*

693 It was noted in Section 5 that the absolute amplitude of the acoustic energy was not
694 the same for all the targets (see Figs. 9a for metals and 10 for other targets). Hence, one
695 could wonder to what extent it depends on target properties or on experimental parameters.
696 Indeed, it was also noted in Section 4 that the pressure, the laser-to-target distance, and also
697 the focus quality play a role in the amplitude of acoustic energy. Measurements that led to
698 results of Section 5 were subject to some variations of these parameters during the course of
699 the experiments that are listed below, and summarized in Table 3:

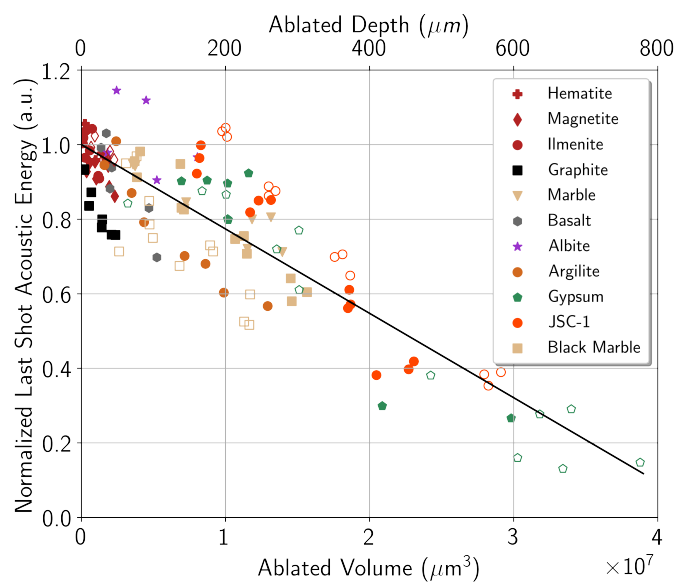


Figure 14: Normalized acoustic energy recorded at the bottom of the laser crater (*i.e.* the mean acoustic energy of the 5 last shots of the burst that created the crater) as a function of the associated crater volume (filled colored markers). Craters were created with 10, 30, 90 and 150 shots. Acoustic energies are normalized by the mean value recorded for the 5 first shots of the sequence. For each target 2 (or 3) craters were created and measured for each number of shots used. The Mars atmosphere results are compared with the same data, extracted from our previous study under Earth atmosphere for the four targets that were used for studies (gypsum, JSC-1, black marble and magnetite; unfilled markers). Mars data are fitted with a linear function $y = 1 - 2.38 \times 10^{-8}x$ (black solid line).

- 700 • Pressure inside the chamber varied from ± 0.3 mbar around 6.22 mbar between the
701 different targets but is precisely known thanks to the pressure sensor.
- 702 • The CWL-based autofocus capability of ChemCam finds the best focus within $\pm 0.5\%$
703 of the real distance. Considering the curves given in Fig. 5, this uncertainty on the
704 focus (corresponding to an uncertainty of 8 mm at this working distance) gives an
705 uncertainty on the acoustic energy of $\pm 20\%$ if we rely on the magnetite and titanium
706 curves or an uncertainty of $\pm 50\%$ for the enstatite. Here we consider $\pm 20\%$ of
707 uncertainty for metal, iron oxides and also graphite that behaves the same way, and \pm
708 50% of uncertainty for all other targets.
- 709 • The laser-to-target distance was changed slightly between targets depending on their
710 thickness. Therefore, a variation of the optical path length of ± 10 mm leads to a
711 variation of the acoustic energy of $\pm 0.5\%$ considering the power law modeled in
712 Fig. 6. This uncertainty is considered negligible compared to the uncertainty on the
713 focus.

714 The amplitude of the acoustic energy is corrected for pressure variations based on the linear
715 relationship determined in Fig. 4. Therefore the uncertainty on the absolute amplitude of the
716 acoustic energy mainly originates from the uncertainty on the focus. The absolute acoustic
717 energy recorded for all the targets and corrected for pressure variations are represented in Fig.
718 15. Error bars correspond to the relative uncertainty on the focus for each target. The figure
719 shows that the graphite sample has a louder sound and its error bar does not overlap other
720 uncertainty intervals. Following graphite, lead and iron have an intermediate acoustic energy
721 with a small uncertainty. All other targets uncertainty intervals intersect the $1 \times 10^{-3} \text{ Pa}^2 \text{ s}$
722 to $2 \times 10^{-3} \text{ Pa}^2 \text{ s}$ acoustic energy range. As for now, the link between absolute value of the
723 acoustic energy and target properties is not reachable with the simple model considered in
724 this work. It should be addressed by upcoming studies.

725 Environmental and instrumental parameters will also vary on Mars, according to the pre-
726 dicted ranges defined in Table 3. The law of change of irradiance with distance for the flight-

727 model of SuperCam was calibrated in laboratory before delivery [6]. The atmospheric pressure
728 will be known precisely, thanks to the Mars Environmental Dynamics Analyzer (MEDA), the
729 weather station of the *Perseverance* rover, with a precision better than 0.05 mbar [75]. Finally,
730 unlike this study, the autofocus capability of SuperCam will rely on the Remote-Micro Imager
731 that provides a precision more than twice as good as the CWL-based autofocus capability
732 [6].

733 Therefore, in order to compare absolute acoustic energies from all the targets on Mars
734 whatever the configuration of their sampling, the following corrections will need to be imple-
735 mented:

736 i Correction for the decrease in irradiance with the distance: Fig 6 showed that acoustic
737 energy is proportional to the irradiance. Acoustic data can be scaled as if the irradiance
738 was the irradiance reached at 1.56 m (distance to the calibration targets).

739 ii Correction for the attenuation of acoustic waves along the sound path length: acoustic
740 data can be scaled as if they were measured at a distance of 1.56 m based on Equation 1
741 and the attenuation coefficient extracted from Bass and Chambers [30]. As the attenua-
742 tion coefficient depends on atmospheric pressure and temperature, *in situ* measurements
743 from MEDA will help to better compute it.

744 iii Correction for background pressure variation. This can be done based on MEDA mea-
745 surements.

746 iv Consideration of the uncertainty associated to the focus. This will provide one term in the
747 assessment of the uncertainties. Based on the value provided in Table 3, this uncertainty
748 will be lower on Mars than during these experiments.

749 **7. Conclusion**

750 Listening to laser-induced sparks produced under Earth atmosphere has shown to provide
751 useful information on target hardness and ablated volume [1]. In an refinement of this work

Variation of Env. and Exp. Parameters	This study	Expected on Jezero Crater, Mars
Pressure	6.22 ± 0.3 mbar	Between 6 mbar and 8.5 mbar depending on the season. Diurnal variations up to $\pm 3\%$ [76]
Laser-to-target Distance	1656 ± 10 mm	From 2.1 m to 7 m and 1.56 m for calibration targets [6]
Focus Quality	$\pm 0.5\%$ of the total distance [52].	$\pm 0.2\%$ of the total distance [6].

Table 3: Variation of environmental (Env.) and experimental parameters (Exp.) that occurred during this study (2nd column) and expected on Mars at the *Perseverance* landing site (3rd column). Variation of air temperature is not listed here as it was not tested in this study but it is expected to vary between 180 K and 260 K depending on the local hour and season [76]. Therefore it will have an impact of air density and sound attenuation coefficient.

752 and in preparation for the SuperCam LIBS investigation on Mars, the acoustic signal from the
753 expansion of the laser plasma on metals, minerals, and rocks, was studied under controlled
754 Mars conditions (carbon dioxide atmosphere and low pressure).

755

756 The sensitivity of the acoustic signal with respect to environmental and instrumental pa-
757 rameters that govern laser ablation and sound propagation was experimentally explored. On
758 the one hand, the amplitude of the acoustic energy increases linearly with the background
759 pressure for the range expected at the Mars 2020 *Perseverance* rover landing site. On the
760 other hand, the laser irradiance at the target controls the intensity of the acoustic signal.
761 The acoustic energy decreases for targets farther to the instrument, due the loss of irradiance
762 with the optical path length (e.g., poorer focusing of the laser beam at longer distances)
763 in addition to a longer distance of attenuation of the sound as it travels back to the in-
764 strument. Furthermore, the sound is louder at best focus, and when the focal point of the
765 telescope is moved away from the target surface, the acoustic energy drops down with the
766 same depth of field as the LIBS optical spectrum intensity. The characterization of the de-
767 pendence of the acoustic energy with respect to these parameters will be used to scale future

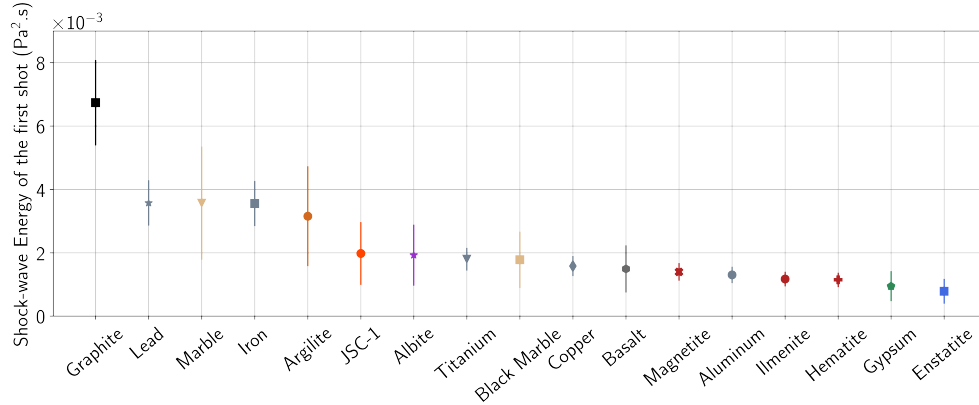


Figure 15: First shot acoustic energy recorded for each target given with error bars that correspond to the relative uncertainty on the focus (see text). All points were scaled to take into account the small difference in background pressure variation between two successive bursts of shots. Targets are sorted by decreasing acoustic energy.

768 Mars data and to compare acoustic signal from targets sampled under multiple configurations.

769

770 The shot-to-shot evolution of the acoustic energy is demonstrated to be intrinsically linked
 771 to the amount of ablated material and the nature of the targeted sample. For metal, the
 772 acoustic energy is almost constant over 150 shots because of a low ablated volume due to a
 773 long thermal penetration depth compared to the optical penetration depth that is responsible
 774 for energy dissipation inside the target. For other rocks and minerals, the shot-to-shot decay
 775 rate of the acoustic energy over a series of laser shots is a decreasing function of the target
 776 hardness. This information will be valuable for the *Perseverance* rover team to estimate the
 777 hardness of potential drill targets and also to track changes of material properties with laser
 778 crater depth, such as in characterizing rock coatings, if they are discovered. The ratio be-
 779 tween the acoustic energy of the first and the last shot of a LIBS burst is seen to be a linear
 780 function of the ablated volume and depth, with the same correlation slope as the one already
 781 observed for ablation under Earth atmosphere. Indeed it was noticed that the ablation rate
 782 under Earth and Mars atmospheres are comparable.

783

784 Finally, this study highlights the potential of the microphone to complement the SuperCam
785 LIBS investigation of rocks and soils by measuring the ablated volume and estimating the
786 target hardness. This work also characterizes the sensitivity of the acoustic energy over
787 a representative range of environmental parameters, instrumental configurations and target
788 properties. The different relationships presented here will help to compare Martian data from
789 one target to another.

790 **Acknowledgments**

791 This work was funded by CNES and Région Occitanie as part of a PhD thesis. We grate-
792 fully acknowledge Guy Perez and Romain Petre Bordenave from CNES for their availability
793 with the 3D-surface profiler. Thanks are due to Jean-Claude Boulliard and the Collection
794 de Minéraux (Sorbonne Université, Paris) for providing mineral samples. The NASA Mars
795 Exploration Program supported some parts (ChemCam body unit, RCW participation).

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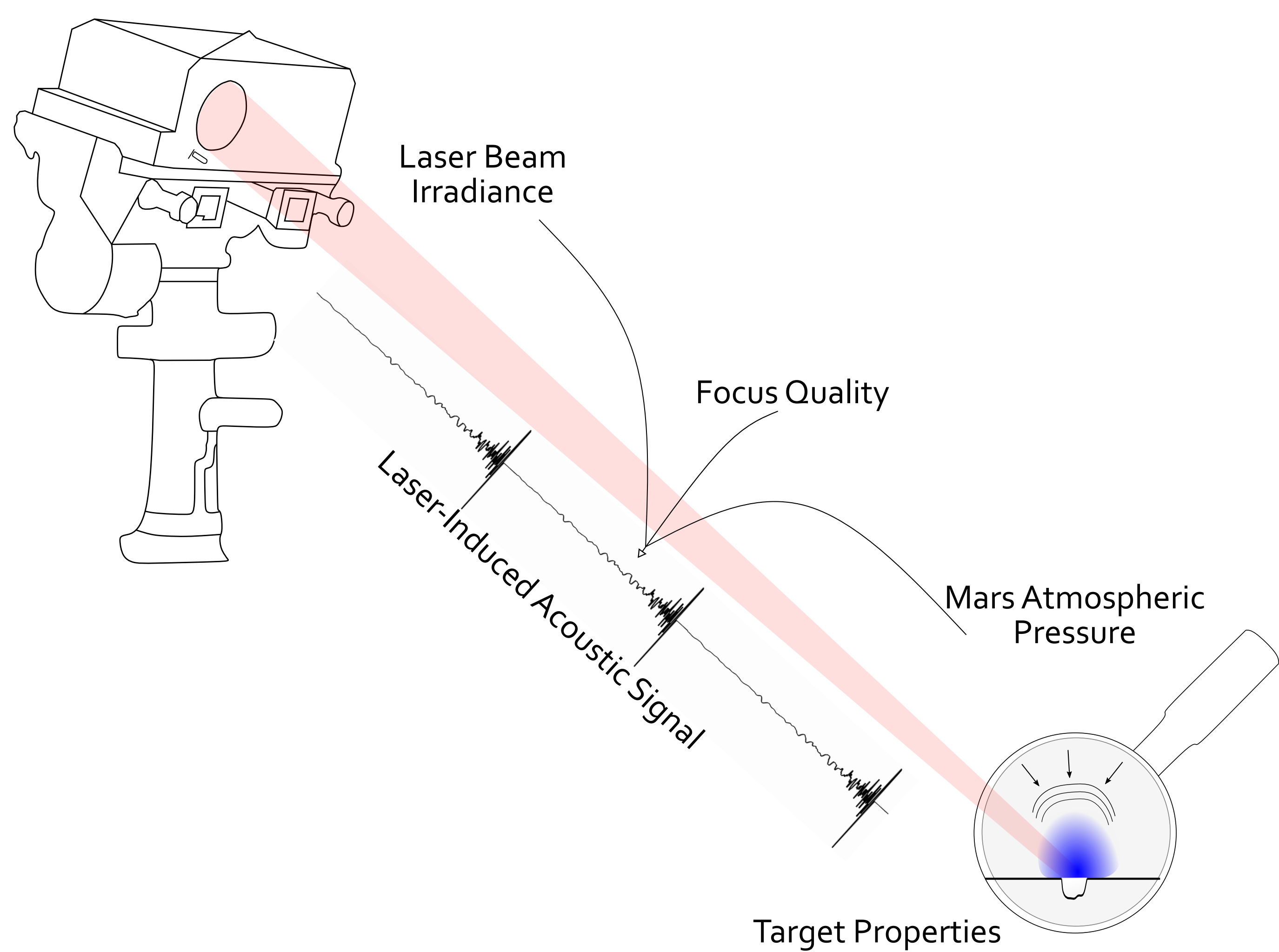
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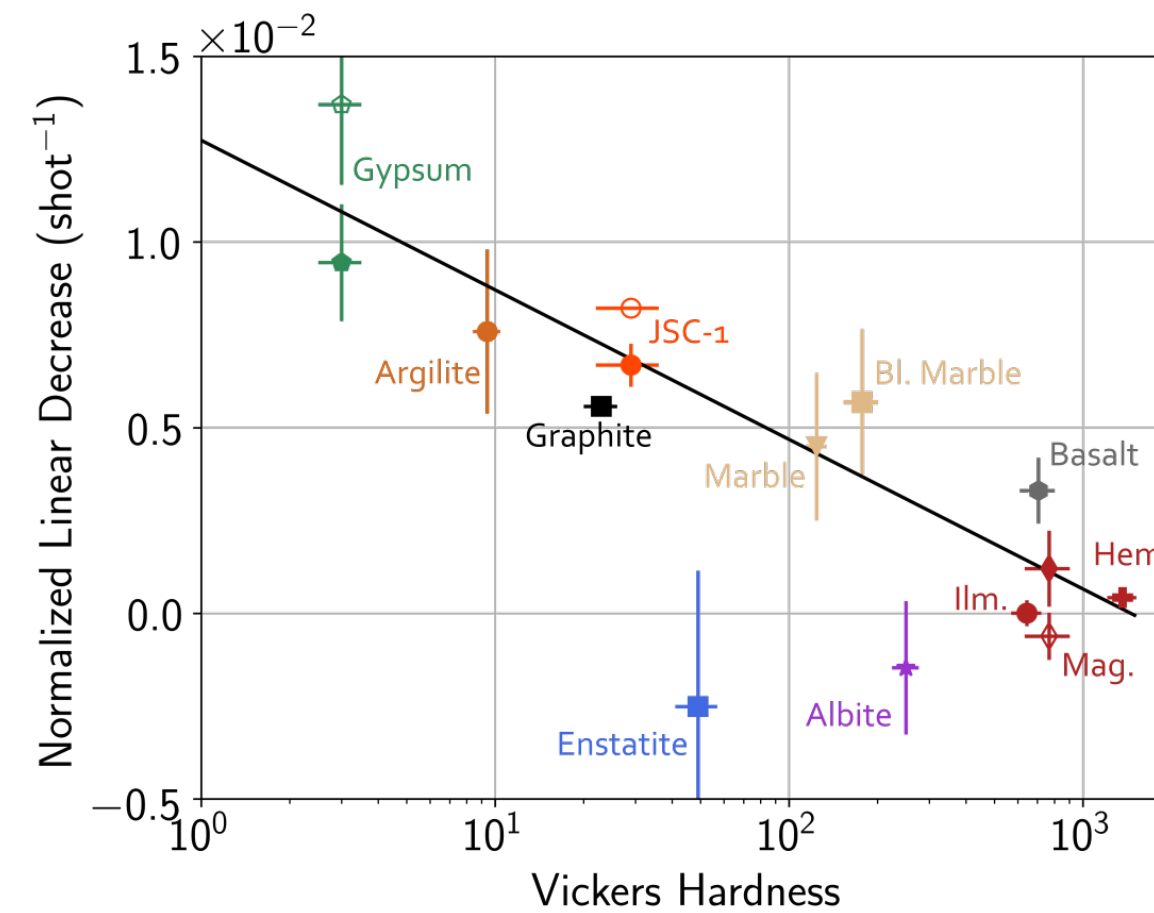
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Recording Laser-Induced Sparks on Mars to...

Estimate target hardness



Measure ablated volume

