Some insight into “bronze quadrigati”: a multi-analytical approach

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Abstract
One of the more obscure areas of Roman Republican early coinage are debased quadrigati, which traditionally are deemed to represent the last stage of this coin type. Starting from a pre-screening, based on low specific gravity samples, we first performed neutron diffraction analyses on a larger sample, which allowed us to single out the more debased series. Subsequently, we focused on those series and applied various additional physical investigations on 18 specimens. The focus of this paper is 17 samples displaying very low silver content. They all belong to a very particular group, refereed as “Apulian” quadrigati in previous literature. They are selected for their numismatic differences and to put in evidence the relevant details of their silver quality, which ranges from “apparently good” to “plain bronze.” In this work, we combine rigorous analytical investigations like X-ray fluorescence (both ordinary and micro-), scanning electron microscope, and neutron diffraction to our accurate numismatic classification of the specimens, leading to a clear correlation between series and debasement. This work aims in particular to gain better insights into these mysterious “silver” emissions, bringing new results that can disclose unknown financial and political facts pertaining to the Second Punic War.

Keywords Ancient coins · Debased quadrigati · Silver surface enrichment · Neutron diffraction · X-ray fluorescence · Scanning electron microscope

Numismatic framework
In this paper, we focus on quadrigati, a pre-denarius silver denomination that was worth two drachms (ca. 6.5 g), produced by Rome at the very beginning of the II Punic War to pay War costs. They were produced at first in Sicily, and in Campania in a later stage; a very small quantity was also produced in Spain, probably in Tarraco, that was the main Roman base for the military operations in Iberia. The production of this denomination ended after the watershed battle of Cannae, in favor of the denarius (Debernardi and Legrand 2014, Walthall 2017, Debernardi and Lippi in preparation). Indeed, current scholarship identifies the root for the denarius birth (Thomsen 1961) in the highly debased quadrigati investigated in this paper. Despite the importance of such a belief, no detailed and systematic study is available so far.

It is relevant to this work to introduce the kinds of adulteration in silver coins of the Roman Republic. There is little doubt that plating is the most important one. A never-ending debate about private or “official” origin of plated coins proves the difficulties of reaching a final statement about this phenomenon (Crawford 1968; Debernardi 2010), and a similar one might arise for the quadrigati considered here. The name of Apulian quadrigati originates from the fact they are almost exclusively found in Apulia, and are at present mostly located in Taranto and Bari museums. Michael Crawford himself defined one of the three hoards (RRCH65) of quadrigati in Taranto Museum as “75 plated quadrigati with legend in relief in linear frame. The hoard is presumably a forger’s stock” (Crawford 1969), while the other two are presented in Debernardi (2016).

Therefore, we need to distinguish clearly between plated and debased coins, which in some circumstances appear very
similar at first sight. A plated coin is a copper or bronze core, covered by a thin highly pure silver foil. A debased coin originates from a core of a copper-silver alloy, where the silver-rich external layer is obtained from applying a blanching procedure called silver surface enhancement (SSE) that preferentially deplete copper from the flan surface, so that the coin appears at sight as pure silver (Debernardi et al. 2017 and reference therein) after the striking.

In this paper, we disclose and quantify different levels of debasement, a topic never addressed before in a systematic way. Of course, it is not the first time that Apulian quadrigati are investigated by analytical techniques, but always in the framework of much broader campaigns, including many coinages and periods. Apart from the book by Hollstein (2000), which is also the most recent, though dating 20 years ago, previous XRF campaigns were carried out by Walker first (1980), and later by Burnett and Hook (1989), on British museum coins, and also by Zwicker (1992). It is useful to present in Fig. 1 Apulian coins analyzed by XRF (X-ray fluorescence) in Hollstein 2000, because they can later be compared with the corresponding series investigated in this paper, by the safer approach of combined analyses presented here.

To make the highest profit of our research, we used an informed screening and sorting of pieces, a topic more specifically pertaining to numismatics. Apulian quadrigati, possibly also for their average bad conditions and scarcity of some of their types, have not received a complete classification by Le Gentilhomme (1934). His work is still the most detailed and it is based on some 130 pieces in the Ailly collection, which, however, does not include all the types of Apulian quadrigati. Numismatic work is in progress to reach a more complete arrangement, but does not depart much from it, except from including rare series not available to Le Gentilhomme. We use in this paper the recently published classification of the Apulian subtypes proposed by Debernardi (2016). However, that paper comprises only the types found in the three Montedoro hoards (Taranto Museum) and in Bari Museum, so the missing (and more debased) types are here introduced for a complete discussion of the Apulian quadrigatus coinage. In Fig. 2, we present, at a glance, all the quadrigati studied with the multi-analytical investigation, covering all the major types. While in this paper we have selected just one non-Apulian quadrigatus, sample Q0, for a multi-analytical analysis, for the sake of completeness, in Appendix 1, we present all the other specimens investigated by neutron diffraction and XRF. This allows for a broader comparison of Apulian quadrigati with other series, with lower degree of debasements and belonging to other mints and times. As shown by the classification of Table 1 (column 1), our sample represents all the nine groups of Apulian emissions, some of them analyzed and defined here for the first time. All of them present a legend in relief, from which their short names stem (Ar = Apulian relief), apart from the unique specimen Q2, displaying an incuse legend (named Ai for this feature). The numbers distinguish the different series (more details in Debernardi 2016), mostly in progression by either weight, specific gravity, or style. This paper will help to decide on a final arrangement of the Apulian series, including the silver content previously unknown.

It is also useful to remind their huge overall amount; from our die counting, we estimate (Esty 2011) about 900 dies, which might have struck some 45 tons of coins, assuming a
standard die-productivity of 10000 coins. These quantities clarify even better the relevance of the present work.

**Measurement strategy and management**

In this paper, we present and compare the results of three measurement techniques: SEM, micro-XRF, and ToF-ND (see Appendix 14 for more details). The Apulian quadrigati are heavily debased coins, which have been subjected to a whitening process by means of the SSE technique. This makes the Apulian quadrigati very complex objects, not at all homogenous as composition and metal texture. The three measurement techniques greatly differ as “investigation-depth” capabilities. SEM accesses just a few micrometers of material below the inspected surface, but provides very clean and precise information on the texture of the inspected area. Micro-XRF goes deeper, but not beyond 100–150 μm from the surface, which is still within the SSE thickness (Ager et al. 2013; Debernardi et al. 2017). Only ToF-ND accesses the whole volume of the coins, but, as a drawback, just provide average values and no information on the spatial distribution of the different components. In several aspects, the techniques are complementary and not self-exclusive, as we
will try to show in the paper. Due to several constraints, like the limited availability of ISIS facility (ToF-ND), accessed when some of the samples were not yet available, not all the samples could be subjected to all the techniques. Only micro-XRF was applied systematically and just for Q15, it was possible to apply the three methods. To compensate for this difficulty, namely the lack of volume data, only accessible via ToF-ND, we developed a strategy of differential analysis of the various parts of the coins, for instance using cracks to access the core of the coin (sample Q2), or even in two cases using a partly destructive method (1-mm edge abrasion) to access the core.

μ-XRF was applied in two different ways: as a scan path (see Fig. 4) or by scanning areas of $300 \times 300 \, \mu m^2$ (on coin surfaces) or $600 \times 600 \, \mu m^2$ on the abraded areas to get average compositional results (see Fig. 3). We followed a consistent policy on all the coins, selecting the cleanest sections in two kinds of regions: “high,” i.e., in correspondence of design reliefs, and “low,” close to reliefs that protect the selected area from wear. This approach is meant to investigate the effects of the strike and of the wear on the outer silvery layer. When possible, we consistently selected the same areas of the horse hind and behind it, like depicted in Fig. 3. When macroscopic traces of silver-enriched layer are present (Q11 and Q12), “low” means the silver spot.

Results and discussions

In this section, we discuss and detail the results presented in summary in Table 1. We sorted the samples per series; in fact, as we will see, samples within each series share similar silver features. Quadrigati were, nominally, silver coins; beyond any appearance they might have today, at the production time, they ought to appear as pure silver pieces. This has to be kept in mind for the following discussion.

Apulian incuse-legend quadrigatus Ai

Sample Q2 belongs to a unique series, since no other Apulian quadrigatus features an incuse legend, as opposed to the legend in relief represented by about one thousand Apulian coins investigated in our numismatic research. It features breaks of its silver-enriched layer, which allow us to get, in just one X-ray brushstroke, both its inner and outer silver content. The results are shown in Fig. 4, where an enlargement of the investigated region shows the X-ray beam path.

The coin was acquired already cleaned, because both the silvery and coppery parts are shiny. In such a case, thanks to the crack of the silver layer, one can see immediately its complex metal structure:

1. A flan made by a 32 wt% silver alloy, here visible in the surface break, was subjected to a surface silver enrichment; the copper in the proximity of the surface was oxidized and eliminated by a blanching bath. This occurs up to a certain depth, and leaves a 95 wt% silver-rich spongy alloy, with poor mechanical features. In fact, what remained after the elimination of the copper is roughly $1/3$ of the whole original alloy volume.
2. The voids left by the copper in the outer layer, after the stroke transferred by the obverse and reverse dies, collapse and form the actual silver-enriched layer.
3. The overall result, in this case, is similar to a plated coin, where the core is not base metal, though. In fact, the silver layer comes from it, via the SSE process, whose details are not described in ancient sources, but reconstructed through experimental archaeology practices (Arles and Téreygeol 2011).

The above procedure is supposedly identical to that used to produce victoriati (Debernardi et al. 2017), but the results are completely different. In fact, the double silver content of victoriati ensures a mechanically much harder SSE layer, which the minting strike is not able to compress in an almost homogeneous layer. This occurs in the present coin, while, in victoriati, the voids are sealed by the thin skin that the strike produces. This strongly modifies the density of victoriati, resulting in reduced specific gravity values, as discussed in our former paper (Debernardi et al. 2017).

Combining the previous discussion with the SG value of $9.40 \, g/cm^3$ for this coin, we directly infer that the silver layer is massive and all the voids are mostly squeezed away. In fact, assuming a perfect binary alloy (which is not the case for this coin), one would get a fineness of $33 \, wt\%$ for this coin, very close to the value of the core alloy of $32 \, wt\%$. However, the SG of the coin provides an average density, which should be higher of that of the only core, due to the higher density of the silver-enriched layer. The small fraction of the total volume$^2$ and the possible presence of minor voids explain the slightly lower measured value.

Apulian relief-legend quadrigati Ar1–Ar2

Series Ar1 and Ar2 are related by style; the former can be seen as the precursor of the latter, displays higher average weight and the emission size is much smaller compared to Ar2, the largest of all the Apulian series, with about 200 identified dies. Sample Q1 belongs to the former, Q3–5 to the latter. Both series display a homogeneous silver quality, at least at sight, for the coins in our database (see Q3 and Q4). Therefore,

$^2$ It might be estimated at about 20% of the whole coin volume (Debernardi 2010), for a crust thickness of 125 μm.
List of the specimens discussed in this paper: samples B1 to B5 refer to specimens from bibliography (Hollstein 2000), depicted in Fig. 1, while samples Q0 to Q17 are depicted in Fig. 2. For each sample, the series and the reference are reported, together with weight (W) and specific gravity (SG). In the right part of the table, the elemental concentration (wt%) is reported, when available, obtained with different techniques: SEM and XRF for the surface content, ToF-ND and XRF on abrasions for the core. For Ag wt% obtained by XRF, more than one value is reported: one per each side of the coin for samples B1 to B5, and two different places for samples Q0 to Q17 (see Fig. 3 for details). The last column reports the Ag-rich phase wt% detected by ToF-ND (see discussion in the text). Sensitivity of SEM analysis is of the order of 0.1 wt% and accuracy around 1%; µXRF sensitivity is tens of ppm, with an accuracy of around 5%. Per their decreasing fineness, the series can be sorted in four groups: Q1, Q2 to Q7, Q8 to Q16, Q17.

| Sample | Series | Reference | W (g) | SG (g/cm³) | Ag (wt%) SEM | Ag (wt%) XRF | Sn (wt%) XRF | Pb (wt%) XRF | Cl (wt%) SEM | Ag (wt%) XRF core | Sn (wt%) XRF core | Ag (wt%) ToF-ND | Phase Ag (wt%) ToF-ND |
|--------|--------|-----------|-------|-------------|--------------|--------------|-------------|-------------|--------------|-----------------|-----------------|-----------------|---------------------|
| B1     | Ar2c   | H 61      | 5.55  | 8.80        | 76            | 73           | Ave         | Ave         | Ave          | Ave             | Ave             | 71              |                     |
| B2     | Ar4    | H 58      | 4.73  | 7.50        | 76            | 85           | Ave         | Ave         | Ave          | Ave             | Ave             | 56              |                     |
| B3     | Ar3b   | H 74      | 4.37  | 8.38        | 55            | 67           | Ave         | Ave         | Ave          | Ave             | Ave             | 32              | 0                   |
| B4     | Ar6    | H 53      | 5.67  | 8.88        | 30            | 31           | Ave         | Ave         | Ave          | Ave             | Ave             | 17              | 3                   |
| B5     | Ar6    | H 73      | 5.94  | 8.68        | 18            | 17           | Ave         | Ave         | Ave          | Ave             | Ave             | 1               |                     |
| Q0     | Dot    | Priv. Coll | 5.32 | 9.60        | 96            | 96           | Ave         | Ave         | Ave          | Ave             | Ave             |                |                     |
| Q1     | Ar1    | Priv. Coll | 6.25 | 9.40        | 85            | 92           | 89          | Ave         | Ave          | Ave             | Ave             | 32              | 0                   |
| Q2     | Ai     | Priv. Coll | 5.82 | 9.40        | 95            | 95           | Ave         | Ave         | Ave          | Ave             | Ave             | 17              | 3                   |
| Q3     | Ar2    | Priv. Coll | 5.32 | 9.15        | 87            | 83           | 85          | Ave         | Ave          | Ave             | Ave             | 33              |                     |
| Q4     | Ar2    | Priv. Coll | 4.67 | 8.90        | 90            | 92           | 91          | Ave         | Ave          | Ave             | Ave             | 29              |                     |
| Q5     | Ar2    | Priv. Coll | 7.47 | 8.89        | 50            | 54           | 52          | Ave         | Ave          | Ave             | Ave             | 37              |                     |
| Q6     | Ar3a   | Priv. Coll | 5.20 | 9.00        | 78            | 64           | 71          | Ave         | Ave          | Ave             | Ave             | 29              |                     |
| Q7     | Ar4    | Priv. Coll | 4.68 | 9.02        | 75            | 89           | 82          | Ave         | Ave          | Ave             | Ave             | 17              | 15                  |
| Q8     | Ar3b   | Priv. Coll | 4.99 | 9.09        | 29            | 40           | 35          | 3           | 1            | 1               | 5               | 9               | 4                   |
| Q9     | Ar5    | Priv. Coll | 5.17 | 8.74        | 34            | 31           | 33          | 5           | 2            | 2               | 1               | 4               | 4                   |
| Q10    | Ar5    | Priv. Coll | 6.40 | 9.00        | 25            | 30           | 28          | 5           | 2            | 4               | 15              | 4               |                     |
| Q11    | Ar5    | Priv. Coll | 4.95 | 8.90        | 18            | 81           | 18          | 5           | Ave          | Ave             | Ave             |                |                     |
| Q12    | Ar7    | Priv. Coll | 4.76 | 8.96        | 32            | 21           | 80          | 21          | 5            | 7               | Ave             |                |                     |
| Q13    | Ar6    | Priv. Coll | 5.67 | 8.95        | 23            | 25           | 39          | 32          | 5            | 2               | Ave             |                |                     |
| Q14    | Ar6    | Priv. Coll | 4.95 | 8.84        | 41            | 25           | 19          | 22          | 6            | 2               | Ave             |                |                     |
| Q15    | Ar6    | Priv. Coll | 5.04 | 8.50        | 28            | 16           | 22          | 10          | 3            | 1               | 14              | 4               | 14                 |
| Q16    | Ar6    | Priv. Coll | 5.40 | 9.02        | 33            | 38           | 36          | 4           | 3            | Ave             | Ave             | 21              | 15                  |
| Q17    | Ar8    | Priv. Coll | 4.77 | 8.67        | 1             | 6            | 6           | 15          | 5            | 3               | Ave             |                |                     |
The new alloy complexity prevents to safely extract from ToF-ND diffractograms the Ag weight percentage, and therefore in Table 1, we provide two columns for Ag: col. 13 for Ag wt%, and col. 14 for the weight percentage of Ag-rich phase. While for the Ag/Cu binary alloys, the phase diagrams are well known, more difficult is the case of ternary alloys as Ag/Cu/Sn. Therefore, we also provide the Ag-rich phase wt% from the ToF-ND analysis, which is the direct result of the measure, and provide the elemental Ag wt% of a corresponding binary alloy. None of the values is the exact Ag wt%, and therefore these results must be carefully handled. As an extreme case, we could comment about Q17, displaying triple amounts of Sn and Pb compared to the previous coins and much lower silver. In this case, if we treat this as a binary alloy, we get 11.7 wt% of silver, but the Ag-rich phase % is zero. This apparent contradiction can be well understood looking at the Ag/Cu phase diagram, where no Ag-rich phase is possible for Ag wt% values below 8.8. Therefore, a value of 0 wt% for this phase just tells that Ag is less than 8.8 wt%, and the corresponding value within a binary alloy framework, rescaled by 0.75 (the total amount of components other than Ag and Cu), fits with this picture. This is also demonstrated by the fact that values of Ag wt% are always higher than those of Ag-rich phase wt%, with an average difference of about 5%.

**Apulian relief-legend quadrigati Ar5 and Ar7**

These series fall within the same low fineness level of Q8, roughly halved compared with the first stage of the production (Ar2, Ar3a, Ar4). For the four coins available in this group (Q9–12), we have the ToF-ND result only for Q9, revealing a fineness < 10 wt%. This sample is however peculiar in several respects. As can be seen in Fig. 5, it represents a different stage/way of surface degradation. In fact, it displays porosities with sizes ranging from 50 to 500 µm (see enlargement in Fig. 5), with some silvery looking together with coppery tones. The reasons of the porous surface remain unclear. μ-XRF results in the two regions (high = chin; low = below chin) confirm that both Ag values are slightly above 30 wt%. This is comparable on average with the surface Q8. These results are important, because they provide the lower alloy fineness, around 9 wt% from ToF-ND data, still compatible with the SSE demonstrated by our investigations.

In Fig. 5, we present for this sample some results of SEM–EDX investigation on the shown area. The Ag-rich phase (in blue) is quite limited at the surface (13.3 wt%); including the silver measured in the Cu-rich phase, surface displays 19 wt% Ag, much lower than 33 wt% from μ-XRF. Corrosion and surface modifications have resulted in a silver depletion from surface in this sample, showing
that SEM–EDX should be applied carefully on such highly debased coins. We also show the investigated areas of Q11–12, the only two samples were we clearly see the former presence of a homogeneous and good silver layer. The difference between these two specimens and Q8–Q10 might reside more in the applied blanching procedure than in the aging process. In fact, in these two specimens, what remains around the silver spots (at around 80 wt% Ag) is much closer to the original alloy composition than in the other three coins: compare the average of 20 wt% with 33 wt% respectively. Clearly the SSE layer features in the two categories are different. In the former case, the alloy to silver transition is much sharper, reminding the features of plated coins, while in the latter case, no sharp transition is found.

To end this group, Q10 remains to be discussed. Different investigations are presented in Fig. 6, for better comparison to Q15, the only other sample we filed on the side (see below). The average fineness detected with μ-XRF on the abraded area is around 15.5 wt%. Also, very useful was the line scan over the abrasion, whose results are shown in the plot. Approaching the surface, we retrieve the well-known transition from core to surface alloy, typical of SSE. In both coins, core composition is reached within 150 μm, a quite typical value also for victoriati (Debernardi et al. 2017). The Ag surface value is almost 40 wt% in this region. It is interesting to observe that core and surface Sn stay almost the same, at about 4 wt%.
This series displays a similar degree of debasement of the previous ones (“6” section). We present as much as four quadrigati belonging to Ar6 group, for they have quite interesting features. In fact, compared to the other series, it seems they have been subjected to a different SSE treatment, which impacts also on their appearance to a visual examination. Two present quite rough, corroded, and copper-like surfaces (Q14 and Q15), while Q13 and Q16 are silvery and possibly quite close to their original appearance.

In Fig. 6, we present SEM and μ-XRF data of specimen Q15. It displays heavy traces of mineral deposits (XRF reveals 15 wt% of silicates) and its overall appearance is that of a bronze coin, no silver or silvering showing up. This sample is the only one subjected to all the analyses described in this paper: ToF-ND, SEM–EDX, μ-XRF, and SEM–EDX applied to the abraded edge. We decided to apply
a destructive approach to avoid any SSE effect; milder abrasions lead to misleading results, as discussed later. Our abrasion of about 1 mm does not bring significant numismatic damage to the coin and is nearly invisible, if the coin is oriented normally; however, it is enough to reach the core of the flan 200 µm below the surface, disclosing its original alloy composition. The SEM image of a portion of the abraded region puts in evidence the coppery and silvery regions (orange and blue).

Silver and copper segregate in separate domains, silver or copper rich, referred respectively as Ag and Cu phases. The average, overall phase composition in this examined area is 12.8% silver, which can be recomputed to a 15 Ag wt% including the silver of the Cu phase. This represents the fineness of the original alloy. Such value is very consistent with the µ-XRF one of 14 wt%. Also, ToF-ND wt% results are consistent, if the binary model is used. Average surface fineness is low, at about 22 wt%, in both Q14 and Q15, which nicely agree with their bronze appearance.

The other two samples have a good silvery look. On the horse hind, where coppery hues can be noticed (see Fig. 7), Q13 displays in fact a dominant Cu phase. Ag-rich phase grains are larger than in the abraded area of Q15, covering 15% of the area, or roughly an overall 23 Ag wt%. This is quite close to 25 wt% values measured by µ-XRF in the same area, which rises to nearly 40 wt% in the less exposed areas, where the silver-rich layer is thicker. These surface results are found nearly the same on Q16, where ToF-ND discloses a slightly higher fineness, in the range 15–20 wt%.

From the previous discussion, it seems that in this series the SSE processes did not guarantee an as bulky silver layer, as in samples Q2, Q11, and Q12, but rather a grainy surface richer in silver. In fact, none of the four surfaces reaches high silver contents; average is 29 wt% and maximum 39 wt% in coin Q13. These values seem in any case sufficient to provide a decent silver looking to the final coins, as shown by the overall appearance of specimens Q13 and Q16. These comments apply also to specimens Q8–Q10, where no silver crust can be found, as opposed to Q11 and Q12.

**Apulian relief-legend quadrigati Ar8**

Ar8 is the last Apulian series and also the smallest one, with its just 12 known specimens, produced by 5 obverse and 7 reverse dies. All of them look bronze coins, as shown by a selection proposed in Fig. 8. This feeling is confirmed by our analyses. In fact, ToF-ND detects no Ag-rich phase for this coin and SEM–EDX just 0.8 wt% Ag. However, µ-XRF values in the two selected areas are quite consistently providing 6.2 and 6.3 wt% Ag. There is no real contradiction in these results. For unknown reasons, there is almost no silver left at the very surface; SEM–EDX map sum spectrum detects a silver content < 1 wt%. Silver is certainly contained in this coin, even though our investigations cannot provide a safe figure of it. In fact, 6 wt% is an average of the first 100 µm, and we do not know the content below that depth.

**SSE layer and its influence in surface investigations**

The condition of many of the selected specimens allowed us to have direct access to the original alloy composition, due to loss, partial or total, of the original silver crust. Surface techniques, like µ-XRF and SEM–EDX, allow also to investigate
very small regions and therefore to differentiate between crust and underlying alloy. This is an advantage of surface techniques, compared to ToF-ND, which cannot detail specific areas. In the “3” section, we have provided a panoramic summary of our investigations, also frequently pointing to the surface layer composition. Clearly, in Apulian quadrigati, the silver added to the alloy was also functional to the final silver looking of the coins. It is surprising how, in most of the cases, such silvering arrived intact to us. In all these cases, only ToF-ND can non-destructively infer the overall fineness, being the outer layer of good silver fineness, at around 85 wt% on averages (Fig. 9). This occurs in almost all the coins belonging to series from Ar1 to Ar4, where the original alloy fineness is found at around 50 (Ar1a only) or 30 wt% (in the other cases). We found high Ag values (at around 80 wt%) also in the SSE of two specimens (Q11 and Q12) belonging to much stronger debased series (at around 15 wt% of silver in the alloy). In all the other cases, however, the outer layer fineness is much lower, averaging at about 30 wt%. We consistently found this phenomenon in just Ar6 series, but also in Ar3b and Ar5, which might point to a different SSE procedure, maybe connected to a different production place.

It is useful to comment about the results on the quadrigati presented in Fig. 2, where XRF was applied with light abrasions of the coin surfaces. It is good to have results from obverse and reverse (see Table 1), which already warns about the limits of the approach. By comparing those results with ours, it appears that in 4 out of 5 coins, they measured the silver content of the SSE layer. The two specimens B4 and B5 are the most interesting to us; in fact, they belong to the series Ar6, which we could investigate with the highest number of specimens (4). Quite remarkably, B5 shares the same dies with Q13 and Q15 the same reverse die with them. We can therefore be nearly sure that all the three coins were produced at the same time (or even day), with the same batch of flans. The TU Clausthal results for these coins differ by almost a factor of 2 (31 wt% vs. 17 wt%). By comparing these with our results, only for coin B5, possibly due to a thin SSE layer combined with abrasion, the results show they reached the core alloy. In our previous paper on victoriati (Debernardi et al. 2017), we were able to find a
calibration factor for SSE modification of the specific gravity, which allows us to infer the coin flan fineness from SG measurements. This might be helpful also for quadrigati, as illustrated in Appendix 3.

**Conclusions**

We presented a detailed investigation, by different and complementary analytical techniques, applied to 18 quadrigati. In particular, we focus on a special class of coins, known as Apulian quadrigati, which are completely separated from the mainstream Roman productions, per style, fabric, weight, and fineness. So far, only sparse analyses were carried out, never lead by a thorough numismatic guidance. One of the novelties of this work is to combine physical and numismatic investigations, which allows to find an order in an otherwise chaotic assembly of data, able so far to detect a debasement, but without a clear framework. Our sample batch is the largest investigated so far and provides guidelines for future and more extensive research.

The achieved data, combined with those from our extensive SG measure campaigns (Debernardi 2016), allows to infer the following debasement pattern for the Apulian quadrigati: a tiny emission at 50 wt% fineness (Ar1a), followed by the bulk of the production (about 80% of the total) at 30 wt% fineness (Ar1b, Ar2, Ar3a, and Ar4). Ar3b, Ar5, Ar6, and Ar7 were produced at around 15 wt%, followed by the ultimate and tiny Ar8, possibly produced at 7.5 wt%. None of the known specimens of this last series presents silvery appearance, and it is not clear what happened to it during the aging and/or circulation process. This series calls for much deeper future investigations.

In summary, we can assume that the fineness of the Apulian Quadrigati might have been subjected to progressive halves: 50, 30, 15, 7.5 wt%.

**Appendix 1: Ag fineness of standard quadrigati**

We provide here the results for less debased quadrigati compared to Apulian ones, which, for their lower debasement, we have not investigated by a multi-analytical approach. Except for specimen Q0, which we selected among these ten pieces as representative for all, revealing good silver at the surface (see Table 1). Combining neutron diffraction, which provides an average silver content, and specific gravity, our model (Debernardi et al. 2017) allows us to estimate the original alloy fineness. Only in this way the results become clear, because different thicknesses of the SSE layer modify both SG and the average ToF-ND fineness. One observes first a small fineness decrease at the end of the quadrigati production, and then a sudden jump by 19 wt% (from 88 to 66 wt% for LG IV A series). Fineness average over 23 victoriati (Debernardi et al. 2017), a denomination immediately following the dot quadrigati in late 216 and produced massively over 4 years, is 68 wt%, demonstrating the tight monetary connection between the two coinages. The 2/3 fineness of all such coins might mark a sudden variation in the Roman Republican monetary system (Debernardi and Lippi in preparation).

**Appendix 2: Analytical methods**

**Neutron diffraction**

Our interest on debased ancient coins is long lasting and resulted in previous publications concerning Cisalpine drachms (Corsi et al. 2015, 2016, 2018) and victoriati (Debernardi et al. 2017), Roman Republican coins almost contemporaneous to quadrigati. In some of those papers, we employed Time of Flight Neutron Diffraction (ToF-ND), one of the most suited non-destructive bulk techniques to infer bulk silver fineness. ToF-ND measurements have been

Table 2: Details of the quadrigati depicted in Fig. 10, reporting classification according to Le Gentilhomme (1934), with presumed mint and date. From the SSE model by Debernardi et al. (2017), we estimate the core, or original flan fineness. The last column averages coins of similar period or mint.

| Coin | Catalogue | Mint | Date       | Weight (cg) | SG g/cm³ | Ag wt% (ToF-ND) | Ag wt% core | Ag wt% time average |
|------|-----------|------|------------|-------------|----------|-----------------|-------------|---------------------|
| LG I-A13 | Sicily | Beginning 216BC | 648 | 9.87 | 93.5 | 88.6 | 89.30 |
| LG I-E4 | Sicily | Beginning 216BC | 677 | 10.08 | 93.2 | 90.0 | 85.53 |
| LG III-1 | Sicily | Mid 216BC | 666 | 10.03 | 89.4 | 86.4 | 85.3 |
| LG I-C | Capua | Beginning 216BC | 646 | 9.85 | 89.4 | 84.9 | |
| LG II-B4 | Capua | Mid 216BC | 634 | 9.85 | 89.9 | 85.3 | |
| LG IV-A | Lilibeum | End 216BC | 640 | 9.68 | 69.5 | 66.5 | 66.66 |
| Q0 | LG IV-A | Lilibeum | End 216BC | 654 | 9.60 | 71.5 | 67.5 | |
| LG IV-A | Lilibeum | End 216BC | 615 | 9.30 | 74.4 | 67.4 | |
| LG IV-B | Tarraco | Beginning 215BC | 615 | 9.65 | 68.8 | 65.6 | |
| LG II-E | Tarraco | Beginning 215BC | 560 | 9.35 | 72.6 | 66.3 | |
carried out at the ISIS pulsed neutron and muon source (Rutherford Appleton Laboratory, UK) using the INES diffractometer. INES is a multipurpose powder diffractometer (Grazzi et al. 2007), often used for archaeometric studies. The entire volume of each coin has been analyzed, adapting the dimensions of the neutron beam using a neutron collimator, a sample aligner, and a neutron camera.

ToF-ND analysis can provide both compositional and structural information. The data obtained consist of a diffraction pattern, showing several peaks whose positions are directly related to the crystal lattice dimensions relative to the different crystallographic phases present in the sample. On silver coins, the analysis of diffraction patterns usually shows peaks related to few phases only. Two major phases have been observed: one is the copper-rich phase (α), while the other is the silver-rich phase (β), as predicted by the biphasic thermodynamic diagram. Then the silver content, which is one of the most important information for numismatists, was calculated using a calibration curve correlating the lattice parameter of the crystallographic phases and the relative Ag/Cu content (Corsi et al. 2016).

For a binary, or about binary, substitutional alloys like the Ag–Cu one relevant in many cases here, one can safely extract the corresponding Ag/Cu ratio. For more complex alloys, containing for instance Sn in relevant amounts like in some of the present samples, one can infer only the Ag-rich phase weight percentage, which is different from the elemental Ag wt%.

**X-ray fluorescence and µ-XRF**

XRF micro-fluorescence was used to obtain compositional data concerning the surface and below the surface of the coins, due to the penetration of the X-rays (in the order of 100–150 µm for a Ag–Cu alloy).

All measurements were performed with the commercial micro-XRF Eagle III-XPL (Roentgenanalytik Systeme GmbH & Co. KG, Taunusstein, Germany), already used for many applications in different fields (Vaggelli and Cossio 2012, Vaggelli et al. 2013, Angelici et al. 2015, Gulmini et al. 2015). The instrument is an elemental analyzer, which combines an optical microscope to an ED-XRF spectrometer. It is equipped with a Rh tube for primary excitation (max voltage 40 kV, max current 1 mA) and a poly capillary lens to focus the X-ray beam onto the sample surface (spot size of 30 µm). The micro-XRF is controlled and operated by the EDAX Vision 32 software, which allows performance of automated spectral acquisition and quantification using single spot acquisition, profiles, or mapping. The quantification programs available rely on the Fundamental Parameters method with standards. The standard used for element calibration are two synthetic glasses, SRM 612 and SRM 610, produced and certified by the American National Institute of Standard and Technology (NIST), each one containing about sixty trace elements with a nominal concentration of 50 and 500 ppm, respectively. The operative conditions are

![Fig. 10 Quadrugati of other series, from private collections, except 7. Museo di Torino, Fabretti 218, characterized by specific gravity and neutron diffraction. Table 2 reports detailed data](image-url)
40 kV, 1000 μA for beam current, 4 μs for time constant, and 500-s lifetime.

**Scanning electron microscope**

This technique accesses the very first hundred nanometers in depth and must be applied very carefully for objects like the coins under investigation. Here we deploy SEM mainly for imaging purposes, which greatly help to access the alloy texture.

Compositional data point analyses were collected using a SEM JEOL IT300LV equipped with an energy-dispersive X-ray spectrometer (EDX), with a SDD (a silicon drift detector from Oxford Instruments), hosted at the Earth Science Department of the University of Turin and already used for different applications (Vaggelli et al. 2019).

It has been used for the determination of major elements at the following working conditions: voltage 15 kV, probe current 5 nA, EDS process time 1 μs, working distance (WD) 10 mm, acquisition time 30 s.

The EDX acquired spectra were corrected and calibrated both in energy and in intensity thanks to measurements performed on cobalt standard introduced in the vacuum chamber with the samples. The Microanalysis Suite Oxford INCA Energy 200, which enables spectra visualization and elements recognition, was employed. A ZAF data reduction program was used for element quantification. The resulting full quantitative analysis was obtained from the spectra, using natural oxides and silicates from Astimex Scientific.

In order to achieve statistically significant counts for a short dwell time, EDS maps were acquired with a short pulse processing time constant. Using a modern silicon drift detector with counting frequencies greater than 100 kcps and selecting a dwell time of 1 ms, an acquisition time of approximately 4 h ($1.4 \times 10^9$ total X-ray counts) for each sample was set.

In the case of average matrix analysis, where the spectrum of each field contains $\approx 5 \times 10^7$ X-ray counts, the analytical precision is better than 0.01 wt %. Each set of X-ray maps was corrected for the instrumental probe current drift due to the long acquisition times by performing an automated measure on a coreference sample of known $(x, y, z)$ coordinates at prefixed 1-h time intervals. For maps, the accelerating voltage was set at 15 kV, the probe current at 2 nA, and the working distance at 10 mm.

**Appendix 3: SG vs. Ag fineness as a function of debasement in Apulian quadrigati**

Related to section “Apulian incuse-legend quadrigatus Ai” and paper by Debernardi et al. (2017), it is interesting to compare in Table 1 the ToF-ND Ag wt% of sample Q0 and Q1. 71 and 56 wt%, with their SG of 9.60 and 9.40 g/cm$^3$ respectively. Those SG values would theoretically correspond to 46 and 33 wt% fineness of a pure Ag/Cu binary alloy, or 73 and 70 wt% for a victoriatus calibrated Ag-SG correspondence, as obtained by Debernardi et al. (2017). Specimen Q0, just preceding of 1 year or less the earliest victoriati, share with them a similar fineness and production steps, so deriving its fineness from its SG by the calibrated relation of (Debernardi et al. 2017) falls within the predicted tolerances ($\pm 6\%$). Instead, sample Q1 features a fineness lower than victoriati (56 wt%), and neither the binary alloy nor the calibrated result (33 and 70 wt%) fits with the actual ToF-ND value. The alloy of this sample lays therefore in the transition region between two regimes: almost binary alloy behavior, below 50 wt% Ag (as for sample Q2), and “victoriatus behavior” for fineness exceeding 65 wt% Ag. Such transition between the two regimes is related to the stiffness of the SSE layer before striking, which is put in evidence here for the first time.

It is therefore useful to display on a single plot all the measured fineness vs. corresponding SG (specific gravity), in order to have in front of us at a glance all the 18 results and have a final discussion about usefulness of SG measures of ancient coins.

![Fig. 11 Silver fineness vs. specific gravity. Crosses, victoriati from Debernardi et al. (2017), dots, results from Table 1 and Table 2. Lines refer to different Ag-SG relations. Blue line, calibrated model for victoriati (Debernardi et al. 2017), with confidence interval by dashed lines; blue circles all belong to the coins in Table 2. The cian line represents an ideal binary alloy. Green and red lines, ideal binary alloys with altered SG values of the individual materials, in such a way to achieve a best fit with the experimental fineness data. For the green curve: $\text{SG}_{\text{low}} = 8.35$ and $\text{SG}_{\text{high}} = 10.70$. For red curve: $\text{SG}_{\text{low}} = 8.2$ and $\text{SG}_{\text{high}} = 16$; they replace $\text{SG}_{\text{Cu}}$ and $\text{SG}_{\text{Ag}}$ in the standard formula for binary alloys.](image-url)
As we had warned (Debernardi et al. 2017), the calibration we achieved works for moderately debased coins, not higher than 65 wt%. However, when measuring SG, one does not know yet the fineness of the coin, as we do in the present case. Therefore, it is worth to exploit the actual results, providing more information on this topic and making SG measurements more useful.

In Fig. 11, we display the fineness vs. SG of the coins in Fig. 3 and Table 1. In the same figure, we report 4 curves, corresponding to relations between SG and fineness for: an ideal binary Ag/Cu alloy (cyan), victoriati (or mildly debased coins, blue), highly and very highly debased coins (green and red). The last two categories comprise all the Apulian quadrigati discussed in this paper. It appears that the Apulian coins split into two families, according to their fineness: debasement in the range 25–55 wt% (green), and below 20 wt% (red). Cleary, SG alone cannot univocally provide the corresponding fineness, as appears from the green and red curves, superimposing in the SG interval 8.8–9.2. This is due to the complexity of the SSE process and its nonlinear dependence from flan debasement and enrichment process. In any case, the best fit presented here might help in providing a rough fineness estimate for the silvery coins, like samples Q1–Q7. Sample Q5 is somehow an exception, because of its high Cl contamination that enhances its greenish patina. However, some bright-silver spots appear on Janus sideburns. If one combines the visual examination with a patina. However, some bright-silver spots appear on Janus sideburns. If one combines the visual examination with a

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Declarations

Conflict of interest The authors declare no competing interests.

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