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# Magnetization curves of sintered heavy tungsten alloys for applications in MRI-guided radiotherapy

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**Purpose:** Due to the current interest in MRI-guided radiotherapy, the magnetic properties of the materials commonly used in radiotherapy are becoming increasingly important. In this note, measurement results for the magnetization (BH) curves of a range of sintered heavy tungsten alloys used in radiation shielding and collimation are presented.

Methods: Sintered heavy tungsten alloys typically contain > 90 % tungsten and < 10 % of a combination of iron, nickel and copper binders. Samples of 8 different grades of sintered heavy tungsten alloys with varying binder content were investigated. Using a Superconducting Quantum Interference Detector (SQUID) magnetometer, the induced magnetic moment m was measured for each sample as a function of applied external field  $H_0$  and the BH curve derived.

**Results:** The iron content of the alloys was found to play a dominant role, directly influencing the magnetization M and thus the nonlinearity of the BH curve. Generally, the saturation magnetization increased with increasing iron content of the alloy. Furthermore, no measurable magnetization was found for all alloys without iron content, despite containing up to 6% of nickel. For two samples from different manufacturers but with identical quoted nominal elemental composition (95% W, 3.5% Ni, 1.5% Fe), a relative difference in the magnetization of 11 - 16% was measured.

**Conclusions:** The measured curves show that the magnetic properties of sintered heavy tungsten alloys strongly depend on the iron content, whereas the addition of nickel in the absence of iron led to no measurable effect. Since a difference in the BH curves for two samples with identical quoted nominal composition from different manufacturers was observed, measuring of the BH curve for each individual batch of heavy tungsten alloys is advisable whenever accurate knowledge of the magnetic properties is crucial. The obtained BH curves can be used in FEM simulations to predict the magnetic impact of sintered heavy tungsten alloys.

Key words: magnetization curve, sintered heavy tungsten alloy, radiation shielding, magnetic fields

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## I. INTRODUCTION

In recent years, the development of new modalities utilising the strengths of MRI devices for the benefit of better radiotherapy treatment has gained substantial interest. Active 50 research in the field of MRI-guided radiotherapy is currently underway, with the first proto types already constructed 1-3. In this process, one major challenge to overcome is the magnetic interference between the MRI device and the hardware components of the radiotherapy treatment system which, historically, were not designed to be operated in a magnetic field. Hence, a characterisation of the magnetic properties of the materials used is essential, 55 i.e. their magnetization or BH curves need to be known in order to predict the magnetic impact. While this information can be readily found in the literature for standard ferromagnetic materials, such as iron, nickel and common alloys of these, there is no publicly available data for the BH curves of so-called sintered heavy tungsten alloys. These alloys typically contain > 90 % tungsten (by weight); the remaining < 10 % are made up by copper, iron 60 and nickel which serve as a binder matrix to increase ductility and machinability<sup>4</sup>. Due to their unique combination of high density, mechanical strength, good machinability and non-toxicity<sup>5</sup>, they are widely used for radiation shielding and collimation purposes (e.g. in MLC's). In this note, the magnetization curves of eight grades of commercially available sintered heavy tungsten alloys have been determined experimentally with a SQUID 65 magnetometer.

## **II. METHODS AND MATERIALS**

## II.A. Sintered heavy tungsten alloy samples

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Table I lists the eight samples of sintered heavy tungsten alloys the magnetization curves of which were experimentally determined. The samples were provided by the two distributors Midwest Tungsten Service<sup>6</sup> and Wolfmet<sup>7</sup>. The selected grades differed in elemental composition, with an iron content varying from 3% down to 0%. The three iron-free samples contained between 6 % and 3.5 % of the relatively less-ferromagnetic nickel. The  $F_x N_x$ sample was cut from a decommissioned Varian Millennium 120 MLC leaf. Although the exact elemental composition was unknown, obtaining the BH curve of this sample was of 75 particular interest due to its widespread use in MLC's. The two samples referred to as

| Sample            | Elem. | compo | Grade |     |                    |
|-------------------|-------|-------|-------|-----|--------------------|
|                   | W     | Fe    | Ni    | Cu  |                    |
| $F_x N_x$         | _     | _     | _     | _   | MLC                |
| $F_{3.0}N_{7.0}$  | 90.0  | 3.0   | 7.0   | 0.0 | $MT17F^{1}$        |
| $F_{1.5}N_{3.5}$  | 95.0  | 1.5   | 3.5   | 0.0 | $MT18F^{1}$        |
| $F_{1.5}N'_{3.5}$ | 95.0  | 1.5   | 3.5   | 0.0 | $\mathrm{HE}395^2$ |
| $F_{0.9}N_{2.1}$  | 97.0  | 0.9   | 2.1   | 0.0 | $MT185^1$          |
| $F_{0.0}N_{6.0}$  | 90.0  | 0.0   | 6.0   | 4.0 | $MT17C^{1}$        |
| $F_{0.0}N_{4.0}$  | 95.0  | 0.0   | 4.0   | 1.0 | $HA195^2$          |
| $F_{0.0}N_{3.5}$  | 95.0  | 0.0   | 3.5   | 1.5 | $MT18C^{1}$        |

TABLE I List of sintered heavy tungsten alloy samples examined in this study. Grades marked with  $^{1}$  or  $^{2}$  were provided by Midwest Tungsten Service<sup>6</sup> or Wolfmet<sup>7</sup>, respectively.

 $F_{1.5}N_{3.5}$  and  $F_{1.5}N'_{3.5}$  were quoted with identical nominal composition, but were produced by different manufacturers. Comparing the results for these samples will indicate if knowledge of the nominal elemental composition (as quoted by the manufacturer) is sufficient to predict the magnetization curve of a sample with respect to a measured reference curve with the same quoted nominal composition. In preparation for the experiments, the samples were cut into cubic blocks of  $3 \times 3 \times 3 \text{ mm}^3$  to fit into the loader cup of the measurement device.

#### II.B. Measurement of magnetic moment

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sign) was used to determine the magnetic moment m of the sintered heavy tungsten alloy samples. The measurements were carried out at a temperature of 300 K. Starting from a fully demagnetized sample, the magnetometer measured the magnetic moment m induced in the sample of the sintered heavy tungsten alloy as a function of applied external field  $H_0$  in the range of 0 to  $8 \times 10^5$  A/m. Data points were acquired with a step width of  $6 \times 10^3$  A/m in the low-field range from 0 to  $90 \times 10^3$  A/m; the step width was increased to  $40 \times 10^3$  A/m

A SQUID magnetometer (Magnetic Property Measurement System 5XL, Quantum De-

<sup>90</sup> In the low-field range from 0 to  $90 \times 10^3$  A/m; the step width was increased to  $40 \times 10^3$  A/m and  $80 \times 10^3$  A/m above  $90 \times 10^3$  A/m and  $240 \times 10^3$  A/m, respectively. The magnetization curve of the MLC-leaf sample had already been determined in a related research project where the step widths in the measurement sequence were slightly different<sup>8</sup>. Since sampling the magnetization curves with the exact same point density was not necessary for this work, the previously obtained data was used (see Table II in the supplemental data).

#### II.C. Data analysis

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The magnetic moment m was normalized by the sample volume, yielding the volumeindependent magnetization M. In materials of finite length, magnetic poles are generated near the ends of the sample which gives rise to a demagnetizing field  $H_d$  opposing the applied field  $H_0^9$ :

$$H_d = N_d M,\tag{1}$$

where the constant of proportionality  $N_d$  is called demagnetizing factor. For the demagnetizing factor, the approximate expression  $N_d = 1/(2n + 1)$  for a uniformly magnetized rectangular rod was used<sup>10</sup>. The dimensional ratio n was 1 for our cubic samples. Subtraction of the demagnetizing field  $H_d$  from the applied field  $H_0$  yields the internal field

$$H = H_0 - H_d. (2)$$

Then, the magnetic flux density B was derived according to the fundamental relation

$$B(H) = \mu_0(H+M),$$
 (3)

where  $\mu_0$  is the vacuum permeability<sup>9</sup>.

#### <sup>110</sup> II.D. Error analysis

The SQUID magnetometer provides the values for the applied magnetic field with a relative accuracy of  $\sigma_H = 10^{-7}H$ . Furthermore, the relative sensitivity for the measurement of the magnetic moment is quoted by the manufacturer as  $\sigma_m = 10^{-4}m^{-11}$ . The samples were cut into cubic blocks with 3.00 mm edge length by the University of Wollongong workshop with an accuracy of  $\pm 0.05$  mm. In order to reduce the measurement uncertainty, the dimensions of each tungsten sample have been measured with calipers to an uncertainty in the edge lengths l of  $\sigma_l = 0.01$  mm. Hence, the uncertainties in the magnetisation M and magnetic flux density B could be derived as

$$\sigma_M = \sqrt{\left(\frac{\partial M}{\partial l}\sigma_l\right)^2 + \left(\frac{\partial M}{\partial m}\sigma_m\right)^2} \approx \frac{\partial M}{\partial l}\sigma_l = 0.01M\tag{4}$$

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$$\sigma_B = \sqrt{\left(\frac{\partial B}{\partial M}\sigma_M\right)^2 + \left(\frac{\partial B}{\partial H}\sigma_H\right)^2} \approx \frac{\partial B}{\partial M}\sigma_M = 0.01\mu_0 M,\tag{5}$$

where the two neglected terms associated with  $\sigma_m$  and  $\sigma_H$  were more than 10<sup>2</sup> times smaller compared to the dominant terms associated with  $\sigma_l$  and  $\sigma_M$ , respectively. Essentially, the accuracy of the experimental data is limited by the uncertainty in the edge lengths.

#### 125 III. RESULTS AND DISCUSSION



FIG. 1 Measured M(H) and B(H) curves of eight different sintered heavy tungsten alloys. The saturation magnetization increases with the iron content of the alloys. Despite up to 6% nickel content, the samples without iron show no measurable magnetization. The  $F_{1.5}N_{3.5}$  and  $F_{1.5}N'_{3.5}$  samples with identical quoted nominal composition show a relative difference in the magnetization of 11 - 16%. The uncertainties in the measurement data are too small to be visualized, but are disclosed in the Supplemental Data V

The magnetization M measured in the H-field range of  $0 - 8 \times 10^5$  A/m is shown in Fig. 1(a) and the corresponding BH curves in Fig. 1(b). As a general trend in Fig. 1(a), the magnetization M(H) becomes higher as the iron content of the sintered heavy tungsten

alloy increases. For instance, a maximum saturation magnetization of about  $14 \times 10^4 \,\mathrm{A/m}$ 

is reached for the  $F_{3.0}N_{7.0}$  sample. The alloys with 1.5% iron, half the iron of the  $F_{3.0}N_{7.0}$ 130 sample, saturate at around  $6 \times 10^4$  A/m; the sample with 0.9 % iron at around  $2.5 \times 10^4$  A/m. The sample cut from the Varian Millennium 120 MLC leaf shows the steepest initial increase of the magnetization M with H which then flattens out and crosses the  $F_{3.0}N_{7.0}$  curve at  $M \approx 10 \times 10^4 \,\text{A/m}$ , before levelling off at about  $13 \times 10^4 \,\text{A/m}$ . Such a saturation magnetization, seen in the context of the curves of the other alloys, could be a sign of an

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Independent of their respective nickel content, no measurable magnetization was found for any of the three alloys without iron, which is in agreement with the results for a measured W-Ni-Cu heavy tungsten alloy found in the literature<sup>12</sup>. These findings suggest that a nickel content of up to 6% is uncritical with regards to magnetic properties and such alloys may be used in environments with magnetic fields without concern.

iron content somewhere between 1.5% and 3% (probably closer to the latter value).

The two samples with identical quoted nominal elemental composition,  $F_{1.5}N_{3.5}$  and  $F_{1.5}N'_{3.5}$ , follow a similar course, however are not in perfect agreement: A maximum relative difference in M of  $16\,\%$  is observed at low H which levels off at about  $11\,\%$  in the regime of magnetic saturation. This discrepancy relates to a maximum difference of 9%145 in the initial section of the corresponding BH curves (Fig. 1(b)). Above magnetic saturation, B and H are linked through a linear relation with a slope of  $\mu_0$  (Eq. 3) and hence the absolute difference between the curves remains constant. The difference can be expressed through the difference in the saturation magnetizations of both samples and amounts to  $\Delta B_{sat} = \mu_0 \Delta M_{sat} \approx 6 \,\mathrm{mT}$ . Considering an uncertainty in the magnetisation of 150  $\sigma_M = 1\% M$  (as discussed in Section II.D), it can be ruled out that the observed difference in the range of 11 - 16% M is a measurement artefact.

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There are a number of factors that could explain the measurement differences. The actual elemental compositions of the two samples could be slightly different from what is quoted since the manufacturers work to a relatively broad tolerance and some variation in the composition between batches is likely. The different sintering procedures used by the manufacturers may have an influence on the magnetic properties by affecting the size and local concentration of the magnetic ions within the alloy. In addition, the grain structure of the constituent metal particles as well as unintentional annealing following the actual sintering may also play a part. In the literature, differences in the magnetization curves 160

for two samples with identical elemental composition (but one in as-sintered, the other in cold-worked condition) have been reported<sup>13</sup> and support this claim. No further steps were undertaken to identify how big a role each of these factors plays in the examined samples as this was not within the scope of this work. However, regardless of which is the dominant factor, the observed difference makes clear that, whenever accurate knowledge of the magnetic properties is crucial, a reference BH curve for an alloy with the same quoted nominal composition may not be sufficient to predict the magnetic impact of sintered heavy tungsten alloys from other manufacturers or even another batch of the same manufacturer. Instead, BH curves should ideally be measured for each individual batch of heavy tungsten alloys in such cases.

IV. CONCLUSION

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Through the measurement of the magnetization curves of eight sintered heavy tungsten alloys, it could be shown that the magnetic properties of sintered heavy tungsten alloys strongly depend on the iron content of the alloys, whereas the addition of up to 6% of nickel <sup>175</sup> in the absence of iron led to no measurable effect. Furthermore, the observed difference in the BH curves for two samples with identical nominal composition but from different manufacturers indicates that the BH curve should be measured for each individual batch of heavy tungsten alloys whenever accurate knowledge is crucial. Using these BH curves as input for FEM simulations to predict the magnetic impact of sintered heavy tungsten alloys will be a great help in the development of new MR-based imaging and treatment modalities in radiotherapy.

V. SUPPLEMENTAL DATA

See Table II below for the measurement data.

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## TABLE II Data of the measured BH curves.

#### $(a)F_{3.0}N_{7.0}$

| $\begin{array}{ll} H\left(\frac{A}{m}\right) & B\pm\sigma_B \ ({\rm mT}) \\ 0 & 0.00\pm<0.01 \\ 5811 & 19.22\pm0.35 \\ 11909 & 35.78\pm0.62 \\ 17900 & 50.33\pm0.82 \end{array}$ |  |
|--|--|
| $\begin{array}{ll} 0 & 0.00 \pm < 0.01 \\ 5811 & 19.22 \pm 0.35 \\ 11909 & 35.78 \pm 0.62 \\ 17900 & 50.33 \pm 0.82 \end{array}$   |  |
| $\begin{array}{cccc} 5811 & 19.22 \pm 0.35 \\ 11909 & 35.78 \pm 0.62 \\ 17900 & 50.33 \pm 0.82 \end{array}$  |  |
| $\begin{array}{ll} 11909 & 35.78 \pm 0.62 \\ 17900 & 50.33 \pm 0.82 \end{array}$   |  |
| $17900  50.33 \pm 0.82$  |  |
|  |  |
| $24007  64.17 \pm 1.01$  |  |
| 29989 $77.02 \pm 1.17$   |  |
| $35961$ $89.35 \pm 1.31$   |  |
| 42015 $101.41 \pm 1.44$  |  |
| 47987 $112.96 \pm 1.56$  |  |
| 53942 $124.18 \pm 1.67$  |  |
| 59968 $135.25 \pm 1.77$  |  |
| $65896$ $145.91 \pm 1.87$  |  |
| 71904 156.49 $\pm$ 1.96  |  |
| 77877 $166.83 \pm 2.04$  |  |
| 83885 $177.04 \pm 2.12$  |  |
| $89993$ $187.19 \pm 2.20$  |  |
| $119963  234.98 \pm 2.50$  |  |
| $159810  295.03 \pm 2.79$  |  |
| 199801 $351.51 \pm 2.98$   |  |
| $239711  405.82 \pm 3.10$  |  |
| $239720  405.67 \pm 3.09$  |  |
| $319620$ $511.05 \pm 3.24$   |  |
| $399566  614.07 \pm 3.32$  |  |
| 479448 715.97 $\pm$ 3.36   |  |
| 559313 $817.40 \pm 3.39$   |  |
| $639313  918.69 \pm 3.42$  |  |
| 719330 $1019.81 \pm 3.43$  |  |
| 799204 1120.63 $\pm$ 3.45  |  |

| (b) $F_{1.5}N_{3.5}$        |                                |  |  |  |
|-----------------------------|--------------------------------|--|--|--|
| $H\left(\frac{A}{m}\right)$ | $B \pm \sigma_B (\mathrm{mT})$ |  |  |  |
| 0                           | $0.00 \pm < 0.01$              |  |  |  |
| 5901                        | $19.55 \pm 0.36$               |  |  |  |
| 11981                       | $33.35 \pm 0.55$               |  |  |  |
| 17972                       | $44.37 \pm 0.65$               |  |  |  |
| 24052                       | $54.63 \pm 0.73$               |  |  |  |
| 30034                       | $64.19 \pm 0.79$               |  |  |  |
| 36006                       | $73.38 \pm 0.84$               |  |  |  |
| 42051                       | $82.44 \pm 0.88$               |  |  |  |
| 48023                       | $91.17 \pm 0.92$               |  |  |  |
| 53978                       | $99.74 \pm 0.95$               |  |  |  |
| 60004                       | $108.28 \pm 0.98$              |  |  |  |
| 65941                       | $116.62 \pm 1.01$              |  |  |  |
| 71931                       | $124.91 \pm 1.03$              |  |  |  |
| 77904                       | $133.12 \pm 1.05$              |  |  |  |
| 83912                       | $141.30 \pm 1.07$              |  |  |  |
| 90020                       | $149.57 \pm 1.09$              |  |  |  |
| 119990                      | $189.56 \pm 1.16$              |  |  |  |
| 159837                      | $241.69 \pm 1.22$              |  |  |  |
| 199810                      | $293.30 \pm 1.26$              |  |  |  |
| 239729                      | $344.43 \pm 1.29$              |  |  |  |
| 239729                      | $344.38 \pm 1.29$              |  |  |  |
| 319638                      | $446.16 \pm 1.33$              |  |  |  |
| 399575                      | $547.44 \pm 1.35$              |  |  |  |
| 479457                      | $648.42 \pm 1.37$              |  |  |  |
| 559331                      | $749.22 \pm 1.38$              |  |  |  |
| 639348                      | $850.10 \pm 1.39$              |  |  |  |
| 719366                      | $950.92 \pm 1.40$              |  |  |  |
| 799240                      | $1051.50 \pm 1.41$             |  |  |  |

(1 ) **F** 

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| (c) $F_{1.5}N'_{3.5}$       |                       |  |  |  |
|-----------------------------|-----------------------|--|--|--|
| $H\left(\frac{A}{m}\right)$ | $B \pm \sigma_B (mT)$ |  |  |  |
| 0                           | $0.00 \pm < 0.01$     |  |  |  |
| 5910                        | $21.44 \pm 0.42$      |  |  |  |
| 11990                       | $36.57 \pm 0.65$      |  |  |  |
| 17972                       | $48.01 \pm 0.77$      |  |  |  |
| 24070                       | $58.58 \pm 0.86$      |  |  |  |
| 30043                       | $68.32 \pm 0.92$      |  |  |  |
| 36024                       | $77.69 \pm 0.98$      |  |  |  |
| 42060                       | $86.86 \pm 1.03$      |  |  |  |
| 48032                       | $95.70 \pm 1.07$      |  |  |  |
| 53978                       | $104.34 \pm 1.10$     |  |  |  |
| 60013                       | $112.98 \pm 1.13$     |  |  |  |
| 65932                       | $121.32 \pm 1.16$     |  |  |  |
| 71931                       | $129.70 \pm 1.19$     |  |  |  |
| 77913                       | $137.95 \pm 1.21$     |  |  |  |
| 83921                       | $146.18 \pm 1.23$     |  |  |  |
| 90020                       | $154.47 \pm 1.25$     |  |  |  |
| 119990                      | $194.53 \pm 1.32$     |  |  |  |
| 159837                      | $246.72 \pm 1.38$     |  |  |  |
| 199819                      | $298.44 \pm 1.43$     |  |  |  |
| 239738                      | $349.64 \pm 1.46$     |  |  |  |
| 239747                      | $349.60 \pm 1.46$     |  |  |  |
| 319656                      | $451.54 \pm 1.51$     |  |  |  |
| 399593                      | $553.00 \pm 1.54$     |  |  |  |
| 479475                      | $654.12 \pm 1.56$     |  |  |  |
| 559349                      | $755.02 \pm 1.57$     |  |  |  |

 $855.98 \pm 1.59$  $956.82 \pm 1.60$  $1057.49 \pm 1.60$ 

 $\begin{array}{c} 639367 \\ 719366 \end{array}$ 

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| $(d)F_{0.9}N_{2.1}$         |                                |  |  |  |
|-----------------------------|--------------------------------|--|--|--|
| $H\left(\frac{A}{m}\right)$ | $B \pm \sigma_B (\mathrm{mT})$ |  |  |  |
| 0                           | $0.00 \pm < 0.01$              |  |  |  |
| 5937                        | $16.40 \pm 0.27$               |  |  |  |
| 12008                       | $26.64 \pm 0.34$               |  |  |  |
| 17981                       | $35.57 \pm 0.39$               |  |  |  |
| 24070                       | $44.24 \pm 0.42$               |  |  |  |
| 30043                       | $52.51 \pm 0.44$               |  |  |  |
| 36015                       | $60.62 \pm 0.46$               |  |  |  |
| 42051                       | $68.71 \pm 0.47$               |  |  |  |
| 48023                       | $76.63 \pm 0.48$               |  |  |  |
| 53969                       | $84.46 \pm 0.50$               |  |  |  |
| 59995                       | $92.34 \pm 0.50$               |  |  |  |
| 65914                       | $100.05 \pm 0.51$              |  |  |  |
| 71922                       | $107.85 \pm 0.52$              |  |  |  |
| 77895                       | $115.58 \pm 0.53$              |  |  |  |
| 83903                       | $123.33 \pm 0.53$              |  |  |  |
| 90002                       | $131.17 \pm 0.54$              |  |  |  |
| 119999                      | $169.59 \pm 0.56$              |  |  |  |
| 159828                      | $220.32 \pm 0.58$              |  |  |  |
| 199810                      | $271.05 \pm 0.59$              |  |  |  |
| 239729                      | $321.59 \pm 0.6$               |  |  |  |
| 239747                      | $321.58 \pm 0.6$               |  |  |  |
| 319647                      | $422.57 \pm 0.62$              |  |  |  |
| 399584                      | $523.42 \pm 0.63$              |  |  |  |
| 479475                      | $624.12 \pm 0.64$              |  |  |  |
| 559331                      | $724.71 \pm 0.65$              |  |  |  |
| 639348                      | $825.45 \pm 0.66$              |  |  |  |
| 719348                      | $926.14 \pm 0.66$              |  |  |  |
| 799276                      | $1026.72 \pm 0.66$             |  |  |  |

## (e) $F_{0,0}N_{c,0}$

| (e) $F_{0.0}N_{6.0}$        |                                | (1                          | $(f)F_{0.0}N_{4.0}$            |                             | $(g)F_{0.0}N_{3.5}$   |                             | $(h)F_xN_x$                    |  |
|-----------------------------|--------------------------------|-----------------------------|--------------------------------|-----------------------------|-----------------------|-----------------------------|--------------------------------|--|
| $H\left(\frac{A}{m}\right)$ | $B \pm \sigma_B (\mathrm{mT})$ | $H\left(\frac{A}{m}\right)$ | $B \pm \sigma_B (\mathrm{mT})$ | $H\left(\frac{A}{m}\right)$ | $B \pm \sigma_B (mT)$ | $H\left(\frac{A}{m}\right)$ | $B \pm \sigma_B (\mathrm{mT})$ |  |
| 0                           | $0 \pm < 0.01$                 | 0                           | $0.00 \pm < 0.01$              | 0                           | $0.00 \pm < 0.01$     | 0                           | $0.00 \pm < 0.01$              |  |
| 5901                        | $7.42 \pm < 0.01$              | 5829                        | $7.32 \pm < 0.01$              | 5892                        | $7.40 \pm < 0.01$     | 3904                        | $16.31 \pm 0.34$               |  |
| 11981                       | $15.06 \pm < 0.01$             | 11918                       | $14.97 \pm < 0.01$             | 11972                       | $15.05 \pm < 0.01$    | 7978                        | $33.25 \pm 0.70$               |  |
| 17963                       | $22.58 \pm < 0.01$             | 17909                       | $22.51 \pm < 0.01$             | 17954                       | $22.57 \pm < 0.01$    | 11990                       | $48.56 \pm 1.00$               |  |
| 24052                       | $30.23 \pm < 0.01$             | 23998                       | $30.16 \pm < 0.01$             | 24052                       | $30.23 \pm < 0.01$    | 12071                       | $48.78 \pm 1.01$               |  |
| 30034                       | $37.74 \pm < 0.01$             | 29989                       | $37.69 \pm < 0.01$             | 30025                       | $37.74 \pm < 0.01$    | 24025                       | $80.69 \pm 1.51$               |  |
| 36006                       | $45.25 \pm < 0.01$             | 35997                       | $45.24 \pm < 0.01$             | 35997                       | $45.25 \pm < 0.01$    | 35979                       | $104.85 \pm 1.79$              |  |
| 42051                       | $52.85 \pm < 0.01$             | 42006                       | $52.79 \pm < 0.01$             | 42051                       | $52.86 \pm < 0.01$    | 48032                       | $126.53 \pm 1.99$              |  |
| 48023                       | $60.35 \pm < 0.01$             | 47978                       | $60.29 \pm < 0.01$             | 48014                       | $60.35 \pm < 0.01$    | 59977                       | $146.41 \pm 2.13$              |  |
| 53978                       | $67.84 \pm < 0.01$             | 53933                       | $67.78 \pm < 0.01$             | 53960                       | $67.83 \pm < 0.01$    | 71922                       | $165.34 \pm 2.25$              |  |
| 60004                       | $75.41 \pm < 0.01$             | 59968                       | $75.36 \pm < 0.01$             | 59995                       | $75.41 \pm < 0.01$    | 71976                       | $165.25 \pm 2.24$              |  |
| 65923                       | $82.85 \pm < 0.01$             | 65896                       | $82.81 \pm < 0.01$             | 65923                       | $82.87 \pm < 0.01$    | 108009                      | $218.92 \pm 2.50$              |  |
| 71931                       | $90.40 \pm < 0.01$             | 71895                       | $90.35 \pm < 0.01$             | 71922                       | $90.41 \pm < 0.01$    | 143826                      | $268.92 \pm 2.65$              |  |
| 77913                       | $97.91 \pm < 0.01$             | 77877                       | $97.87 \pm < 0.01$             | 77895                       | $97.91 \pm < 0.01$    | 179778                      | $317.48 \pm 2.75$              |  |
| 83921                       | $105.47 \pm < 0.01$            | 83903                       | $105.44 \pm < 0.01$            | 83930                       | $105.50 \pm < 0.01$   | 215830                      | $365.27 \pm 2.82$              |  |
| 90029                       | $113.14 \pm < 0.01$            | 89993                       | $113.09 \pm < 0.01$            | 90011                       | $113.14 \pm < 0.01$   | 251737                      | $412.22 \pm 2.88$              |  |
| 120008                      | $150.82 \pm < 0.01$            | 119972                      | $150.77 \pm < 0.01$            | 120008                      | $150.85 \pm < 0.01$   | 287707                      | $458.89 \pm 2.92$              |  |
| 159855                      | $200.89 \pm < 0.01$            | 159828                      | $200.86 \pm < 0.01$            | 159855                      | $200.93 \pm < 0.01$   | 323767                      | $505.37 \pm 2.96$              |  |
| 199837                      | $251.14 \pm < 0.01$            | 199819                      | $251.11 \pm < 0.01$            | 199828                      | $251.18 \pm < 0.01$   | 359710                      | $551.60 \pm 2.99$              |  |
| 239756                      | $301.30 \pm < 0.01$            | 239729                      | $301.27 \pm < 0.01$            | 239747                      | $301.36 \pm < 0.01$   | 395662                      | $597.53 \pm 3.01$              |  |
| 239765                      | $301.31 \pm < 0.01$            | 239738                      | $301.27 \pm < 0.01$            | 239747                      | $301.36 \pm < 0.01$   | 799258                      | $1109.87 \pm 3.16$             |  |
| 319674                      | $401.74 \pm < 0.01$            | 319638                      | $401.68 \pm < 0.01$            | 319665                      | $401.81 \pm < 0.01$   |                             |                                |  |
| 399611                      | $502.19 \pm < 0.01$            | 399575                      | $502.14 \pm < 0.01$            | 399602                      | $502.29 \pm < 0.01$   |                             |                                |  |
| 479493                      | $602.58 \pm < 0.01$            | 479466                      | $602.54 \pm < 0.01$            | 479484                      | $602.70 \pm < 0.01$   |                             |                                |  |
| 559349                      | $702.93 \pm < 0.01$            | 559331                      | $702.90 \pm < 0.01$            | 559349                      | $703.09 \pm < 0.01$   |                             |                                |  |
| 639366                      | $803.49 \pm < 0.01$            | 639348                      | $803.46 \pm < 0.01$            | 639366                      | $803.67 \pm < 0.01$   |                             |                                |  |
| 719384                      | $904.05 \pm < 0.01$            | 719366                      | $904.02 \pm < 0.01$            | 719402                      | $904.28 \pm < 0.01$   |                             |                                |  |
| 700259                      | 1004 49 < 0.01                 | 700050                      | $1004.41 \mid < 0.01$          | 700050                      | 1004 65 1 < 0.01      |                             |                                |  |

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