三维空间结构叠加单原子成份信息的四维成像

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摘要：透射电子显微镜（TEM）和原子探针层析技术（APT）是目前最先进的两种显微结构表征技术，各有千秋：TEM 擅长形貌和晶体学结构分析，APT 以揭示单原子成份信息见长。理论上，两种技术可以优势互补，将 APT 的结果叠加到 TEM 结果中，可以完美解决 TEM 对游离态原子无法表征的难题，也可以解决 APT 结果中晶体学结构信息缺失的问题。但现实是，由于 APT 结果在三维空间上存在严重的局部畸变，难以与“眼见为实”的 TEM 结果在空间上直接匹配。尽管两种技术都拥有原子尺度的分辨极限，但获得的结果往往难以互恰，给研究者们带来极大的困扰。本研究基于这一背景，通过优化 APT 纳米针尖试样的制样设备和方法，改制 TEM 样品杆，用纳米针尖试样取代 TEM 传统的薄片状试样，试样在完成 TEM 观察之后直接转移到 APT 设备中“原位”表征，利用 TEM 结果中的特征界面为 APT 结果的三维重构提供参考系校正局部畸变，从而实现 TEM 结果与 APT 结果互恰。本研究为贵重设备的二次技术开发和设备互关联提供了很好的范例，也为老旧设备通过技术革新实现“枯树新枝又一春”提供了新思路。

关键词：透射电子显微镜；原子探针层析技术；三维重构；析出相；晶界

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The four-dimensional imaging with integration by bringing monatomic composition into the three dimensional spatial structure

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Abstract: As the two most advanced characterization techniques, transmission electron microscopy (TEM) and atom probe tomography (APT) have their merits: TEM is skilled in

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revealing the microstructural morphology and crystallography structure of materials, and APT is good at detection of elemental composition of matter at atomic level. It seems that they are complementary, the combination of the two can bring the atomic composition distribution into the microstructures observed by TEM. However, in fact, the analysis makes researchers confused in most cases, due to the local distortion in the reconstructed APT result, which makes the microstructural morphology in APT data much difficult to matching that in TEM image. Based on the above background, this project focus on the TEM and APT relating technology. We explored a full-automatic APT sample preparation system and a full-vision TEM holder for APT specimens, which makes it possible to correct the local distortion in APT reconstructed results by introducing the in-situ observation in TEM before APT experiment. Furtherly, we proposed a 4DTEM technique, a combination of TEM and APT, providing the distribution of atomic composition on the TEM image in 3d space. This work shows a successful case for the second development of the large expensive instruments and the intercorrelation of different equipment.

Key words: Transmission electron microscopy; Atom probe tomography; 3D reconstruction; Precipitate; Grain boundary

透射电子显微镜（TEM）和原子探针层析技术（APT）是材料显微结构表征领域目前最先进的两类表征技术[1-6]。TEM 以“眼见为实”著称，电子束“照射”纳米薄片试样，试样内部的微结构会被投影到显示屏上。显然，TEM 眼见的只是材料内部结构的二维投影，对具有周期性结构的晶体而言，可以通过二维投影按照结构的周期性还原原子在空间的三维排列。但对于一些没有周期性结构或周期性结构不明显的组织，如合金化或元素掺杂形成的游离态随机分布的合金元素原子，又如原子排列鲜有规律的大角晶界的结构性质，没有晶体结构的原子团簇，等，TEM 无能为力。

APT 技术是在场离子显微镜基础上发展起来的一种分析技术，可以提供高灵敏度的三维空间成像和化学成份测量，它能探测到浓度为百万分之一的痕量元素，对轻量元素 H、Li、Be、C 等一样适用[7, 8]。APT 采用纳米针尖试样，试样的尖端半径要求<100nm。在超高真空及液氮冷却条件下，在针尖试样尖端施加足够的正高压，形成电场脉冲（有时还会辅助激光脉冲），试样表面原子开始电离并尝试离开试样表面（场发射）。在样品与近局域电极之间施加电场，针尖端表面形成的场蒸发离子将会经过直流电场飞向后端的飞行时间质谱仪和位置敏感探测器。飞行时间质谱仪根据离子的飞行时间，可以推算每个离子的质量/电荷比