Development of an in-situ analysis instrument for microstructure of materials with low temperature

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Abstract. The development of instruments capable of dynamically observing the microstructure at different temperatures is of great significance for the study of materials. The liquid nitrogen and a self-developed high-frequency pulse tube cryocooler were used as the cold source to develop the cryogenic system of scanning electron microscope (SEM) in this paper. The vibration and temperature control problems and solutions involved in using these two cold sources as SEM cryogenic systems were described and discussed in detail. It was found that it is necessary to fully achieve the heat balance at a certain temperature and adjust the image displacement to overcome the adverse effects caused by the thermal expansion and contraction of the low temperature components, and the cryocooler needs to use an intermittent operation mode to avoid distortion of the material microstructure image caused by vibrations of the cryocooler. Whether it is for liquid nitrogen or cryocooler, a thermal switch between the cold source and the thermal bridge is helpful for temperature control of the sample and reduced heat leakage of the system.

Introduction
The development of instruments capable of dynamically observing the microstructure at different temperatures is of great significance for the study of materials [1, 2]. The cold sources of cryogenic scientific instruments mainly include cryogens and cryocoolers. For cryogens, the most used is liquid nitrogen, but its deficiency is that the operating temperature is relatively high (77 K, by reducing the gas pressure, the minimum can be reduced to 63 K). In order to obtain a lower cooling temperature, liquid helium (about 4 K) is required, but the cost will be significantly increased. For cryocoolers, the most widely used ones are GM cryocoolers or GM pulse tube cryocoolers, which have the advantage of achieving very low cooling temperatures (less than 3K) and a large cooling capacity (larger than 1.5W/4.2K), but the large size/weight (>100 kg) and power consumption (7-8 kW) of such cryocoolers limit their application in some applications; There are also reports of using high-frequency cryocooler with smaller size/weight (10-20 kg) and lower power consumption (<0.5 kW) as cold sources for cryogenic scientific instruments [2, 3]. However, such cryocoolers have very small cooling capacity in the same temperature zone, and special attention must be paid to efficient heat transfer and effective heat insulation during the development process to minimize system heat leakage [4, 5].

In this paper, based on a commercial scanning electron microscope (SEM, model SU1510, produced by Hitachi), liquid nitrogen and self-developed high-frequency pulse tube cryocoolers are used as cold sources to develop in-situ observation instruments for material microstructure. The reason for using...
liquid nitrogen as a cold source before using the cryocooler is to investigate the effect of the introduction of the external cooling system on the SEM system, and explore some general methods of vibration and temperature control.

**SEM cryogenic system cooled by liquid nitrogen**

The schematic and photograph of the SEM system using liquid nitrogen as a cold source are shown in Figures 1 and 2, respectively. In fact, the original solution was to fix the liquid nitrogen system on another support shelf, but found that it was necessary to introduce a special vibration reduction device to the support shelf: if without additional vibration reduction device, the image may be distorted due to the ground vibration when the magnification of the observation is high. After adopting the scheme shown in Figures 1 and 2, that is, the liquid nitrogen system is integrated with the SEM’s sample chamber, and the vibration is reduced by the existing vibration reducing device, the original imaging quality of the SEM can be maintained. However, the test results show that during the cooling process, the volume of the sample and other components connected to it (such as the sample holder and the thermal bridge) change due to thermal expansion and contraction, which will cause image shift and thus affect the image quality. Therefore, in order to effectively observe the microstructure of the material, it is necessary to reach the thermal equilibrium at a certain temperature, and then observe the microscopic morphology of the next temperature. Through the stepwise operation of selecting multiple temperature points in the temperature zone to be tested, the dynamic observation of the material’s microstructure at different temperatures can be realized. During the test, it is also necessary to continuously adjust the XYZ displacement adjusting device to overcome the positional deviation of the material due to thermal expansion and contraction (In fact, it will only shift in one direction), thereby realizing the observation of a certain fixed position of the material.

![Figure 1. Schematic of SEM cryogenic system (cooled by liquid nitrogen).](image1)

![Figure 2. SEM cryogenic system photo (cooled by liquid nitrogen).](image2)
For temperature control, in order to be able to control the sample to any temperature between 77 K (the internal pressure of the liquid nitrogen tank is equal to the atmospheric pressure) and room temperature, a thermal switch is required between the liquid nitrogen tank and the thermal bridge. The thermal switch is a gas gap structure that controls the thermal resistance between the liquid nitrogen tank and the thermal bridge to a suitable range by controlling the amount of helium inside. Then the PID was used to control the electric heating of the sample holder to achieve precise control of the sample temperature (Test results show that temperature control accuracy can reach ±0.1 K). Figure 3 is a photomicrograph of the sample holder without sample (oxygen-free copper) at 80 K by SEM. It should be noted that another purpose of controlling the thermal resistance between the liquid nitrogen tank and the thermal bridge through the thermal switch is to reduce the heat leakage between the two while heating the sample holder.

**Figure 3.** A photomicrograph of the sample holder (oxygen-free copper) without sample at 80 K.

**SEM cryogenic system based on high frequency pulse tube cryocooler**

Based on the development of above liquid nitrogen cooling system, further development was carried out using a cryocooler as a cold source. A self-developed high-frequency pulse tube cryocooler was adopted, which can achieve a no-load temperature of less than 5.7 K and provide a cooling power of 80 mW at 8 K or 400 mW at 15 K with an electric power input of 400 W [6]. Figure 4 and Figure 5 are the schematic and cooling capacity curve of the developed high-frequency pulse tube cryocooler, respectively. A prominent feature of pulse tube cryocooler is that there are no moving parts at the low temperature part, which has significant advantages in terms of low vibration and operational reliability. Therefore, the vibration of the pulse tube cryocooler mainly comes from the compressor. In order to reduce vibration, the developed pulse tube cryocooler in this paper uses an oppose-dual linear compressor to reduce vibration by the relative movement of two opposed pistons. In addition, a flexible tube connection is used between the compressor and the cold finger to further eliminate the vibration comes from the compressor. Through testing, it has been found that this vibration reduction scheme can indeed be useful under an optical microscope (tens or hundreds of magnification observations). But for SEM (magnification observations of thousands or even tens of thousands), the cryocooler’s vibration still has a significant impact on the imaging of the SEM, especially when the operating temperature of the cryocooler is very low (the lower the temperature, the greater the electrical power required, resulting in greater piston movement of the compressor), image distortion will occur and it will be completely unobservable. Therefore, some new strategies to reduce vibration are needed. Figures 6 and 7 are schematic and three-dimensional design views of the SEM cryogenic system, respectively. Based on this system, we conducted research on vibration reduction measures, which will be discussed in detail below.
Figure 4. Schematic of the SEM cold source-high frequency pulse tube cryocooler.

Figure 5. Cooling power of the SEM cold source-high frequency pulse tube cryocooler.

Figure 6. Schematic of SEM cryogenic system (cooled by high frequency pulse tube cryocooler).
In order to solve the vibration problem of the cryocooler, we adopted an intermittent working mode: When the sample temperature is higher than the set temperature, start the cryocooler to cool down (high vibration state). When the sample temperature is significantly lower than the set temperature, turn off the cryocooler (no vibration), and maintaining the temperature of the sample at the set temperature by controlling the heating amount of the sample holder by PID (test results show that temperature control accuracy can also reach ±0.1 K), thus, the observation of the microscopic morphology of the sample at this temperature is completed. In fact, during the temperature control, the thermal bridge with a lower temperature than the sample is equivalent to a cold source, which is used to meet the cooling demand of the sample. In addition, the heat bridge has a large heat capacity, so that its temperature rises very slowly, which can provide sufficient time for temperature control of the sample.

It can also be seen from Figure 6 that a thermal switch is also installed between the thermal bridge and the cold head of the cryocooler. However, it is slightly different from the thermal switch function of the liquid nitrogen cooling system, it usually has only two working states (on or off): When the cryocooler is in the start-up state, the heat switch is filled with helium to enhance the heat transfer between the cold head of the cryocooler and the heat bridge; When the cryocooler is in the off state, the helium inside the thermal switch is extracted to reduce the heat transfer between the cold head of the cryocooler and the thermal bridge.

In general, after using the above-mentioned vibration reduction and temperature control methods, the SEM cryogenic system cooled by the cryocooler can work normally, and the microstructure of the material at different temperatures can be observed. However, this cooling system needs further optimization work, and in order to better observe and analyse the materials, it also needs to add some new functions, for example, on the one hand, a deformation excitation mechanism can be added to achieve deformation excitation such as stretching and compression of a material sample at different temperatures; on the other hand, a multi-stage high frequency pulse tube cryocooler capable of achieving a lower temperature (to reach liquid helium temperature range) will be used to further broaden the temperature observation range of the sample [7, 8].

Conclusions
In order to dynamically observe the microstructure of materials at different temperatures, the liquid nitrogen and self-developed high-frequency pulse tube cryocooler were used as the cold source to develop the cryogenic system of SEM. The test results show that coupling the cryogenic system to the SEM sample cavity and sharing the existing vibration reduction device of the sample cavity does not affect the imaging quality of the SEM. However, the sample and other components connected to it will
expand and contract due to temperature changes, which will cause the image to shift and thus affect the image quality. Therefore, it is necessary to reach the thermal equilibrium at a certain temperature, and then observe the microscopic morphology of the next temperature in the process of cooling. Meanwhile, it is also necessary to continuously adjust the XYZ displacement adjusting device to overcome the positional deviation of the material. Whether it is for liquid nitrogen or cryocooler, a thermal switch between the cold source and the thermal bridge is helpful for temperature control of the sample and reduced heat leakage of the system. When the SEM is at a high observation magnification, the cryocooler needs to use an intermittent operation mode to avoid distortion of the material microstructure image caused by the cryocooler’s vibration.

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