Cooling to 10 K on a Four-Circle Diffractometer by means of a Double-Stage Cryorefrigerator

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Abstract
The cooling of single-crystal samples down to 10 K within the Mark VI Eulerian cradle of a neutron four-circle diffractometer has been achieved by means of a two-stage cryorefrigerator fixed to the offset \( \chi \) circle and coupled to the \( \phi \) shaft. There is no blind spot for the \( \phi \) rotation but the \( \chi \) rotation must be restricted to the \(-100 < \chi < 50^\circ\) range to avoid collisions of the cryorefrigerator with the detector shielding. The major advantages of this device are its low running cost and its ease of use, no liquid nitrogen or helium being required.

Introduction
Neutron diffraction experiments at very low temperatures are usually carried out using standard reservoir-type liquid-helium cryostats. However, in the case of elastic scattering of neutrons by single crystals, where the most satisfactory measurement geometry is the four-circle geometry, the complicated movements of an Eulerian cradle are not desirable with such a cryostat. Various solutions have been found to this problem, the lowest temperatures being generally obtained by means of rather sophisticated continuous-helium-flow cooling units. An example is the system capable of 3.5 K proposed by Herbert & Campbell (1977) which we mention because it may also be mounted on the cradle of a Mark VI\( ^\dagger \) diffractometer. Another interesting solution is the use of either a single- or double-stage closed-cycle cryorefrigerator.

Double-stage units have already been installed on single-crystal diffractometers, for example using an unusually large Eulerian cradle (Koetzle & Henriques, 1976), or by means of a sophisticated mechanical adaption including a flexible heat-transfer link as designed by Samson, Goldish & Dick (1980) for conventional X-ray apparatus.

The system described here comprises a commercial, closed-circuit, two-stage cryorefrigerator – a Displex CSA 202 manufactured by Air Products\( ^\ddagger \) – capable of a minimum temperature of 10 K, a special alignable mount and an overhead pulley system for the gas and electrical supply lines. Its appropriate code number would be '643170' for the bibliographic computer code defined by Rudman (1977) for low-temperature diffraction apparatus. Its principal advantages over the other types of system are its very low running cost and simplicity of use, mainly because liquid nitrogen and liquid helium are not needed. The economy thus made in case of intensive use will rapidly cover the cost of purchase.

General description
The cooling unit (Fig. 1) is a closed-cycle cryogenic refrigeration system employing helium gas as a working medium. The system consists of a compressor module with electrical controls, flexible interconnecting gas lines and the expander module in which the refrigeration is produced by means of a Solvay cycle of the helium. The cryorefrigerator is obtainable as a single-stage unit capable of a minimum temperature of 45 K or as a two-stage unit capable of 10 K. The expander of the

Fig. 1. A block diagram of the cooling system: (1) valve motor; (2) first-stage displacer; (3) second-stage displacer; (4) sample; (5) \( \phi \) drive; (6) \( \chi \) circle; (7) low-pressure helium return; (8) high-pressure helium supply; (9) compressor; (10) temperature controller; (11) vacuum pump.

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\( ^\dagger \) Manufactured by Grubb-Parson, Newcastle upon Tyne, England.
\( ^\ddagger \) Air Products and Chemicals, Inc., PO Box 538, Allentown, PA 18105, USA – approximate price in 1979 $7000.
former is small enough to be installed inside the Eulerian cradle of a neutron four-circle diffractometer

(Lehmann, Mason, Simms & Allibon, 1980) while the latter is not. However, for the system described this difficulty does not exist due to the peculiarity of the Mark VI Eulerian cradle which possesses a \( \chi \) circle offset by about 80 mm thus liberating the access from above to the \( \varphi \)-circle drive plate. Hence the cold head of a two-stage cryorefrigerator can be mounted vertically above the \( \varphi \) circle as shown in Fig. 2.

The driving of the assembly poses a few problems. The DC motors used to drive the \( \varphi \) and \( \chi \) circles have ample power but the anti-backlash gears attached to these motors accept only relatively small loads. This is not too important for the \( \chi \) circle since the weight of the assembly is of the same order as the weights normally attached to the circle for counter-balancing the motor/encoder assembly of the \( \varphi \) circle. For the \( \varphi \) circle, however, it was found necessary to use stronger springs for the anti-backlash gears and to reduce as much as possible the 'drag' of the pressure lines, electrical leads, etc. This has been achieved with the aid of an overhead beam and pulley system (Fig. 3) which guides the various cables, by replacing the original gas lines by very flexible high-pressure (20 \( \times \) 10^5 Pa) neoprene tubing and by running these services out from the top of the assembly.

The valve motor housing (H, Fig. 2) of the cold head protrudes some 250 mm from the \( \chi \) circle and there is a...
risk of collision with the detector shield if $\chi > 50^\circ$. For the other angles no additional angular limitations are introduced. The existing limitations are mainly inherent in the offset $\chi$-circle geometry and may be largely reduced by means of some computational effort to find an appropriate azimuthal rotation for each ‘blind’ reflection (Filhol & Thomas, 1976). The useful angular zone is therefore $-100 < \chi < 50^\circ$. To allow for possible false manoeuvres an anti-collision switch was installed. It consists of a flexible metal strip mounted on springs around the detector housing and electrically insulated from it. Contact with any part of the diffractometer or of the cryorefrigerator supplies a signal to stop all drive motors of the diffractometer. In fact, because of inertia, rotations actually stop after a few degrees, the over-run being absorbed by the flexible strip and the springs.

**Use and performance**

The system described (Fig. 2) has been designed to enable easy and rapid sample mounting and alignment. Once the assembly has been installed on the Eulerian cradle the sample mounting is carried out by disconnecting the $\varphi$ drive and sliding up the telescope shields ($D_1$, $D_2$). The inner can ($D_3+D_{31}$), into which the sample is previously mounted, is then screwed onto the cold head of the expander ($G$). This can act as an environmental as well as an isothermal shield.

The alignment of the sample is made possible by means of the translational movements $x$ and $y$ ($C_1$) which provide an adjustment of $\pm 3.5$ mm, and the vertical movement $z$ ($C_2$) with an adjustment of $\pm 6$ mm. Once the sample is mounted and with the shields and $\varphi$ coupling in place, preliminary centering may be carried out by taking photographs in the direct beam using a neutron camera. For this method, crystals which are weak neutron absorbers may be glued onto a cadmium pin. Optical centering of the crystals is also possible using a different mounting procedure but this method is rarely used. In either case, final alignment of the sample is normally made by the usual crystallographic method.

The time needed to cool down the sample to 10 K is approximately two hours once the required vacuum of at least $1.3 \times 10^{-2}$ Pa has been obtained. After this initial pump down and at temperatures lower than 175 K the system, aided by a zeolite container, will cryopump, thus making it possible to valve off the vacuum line and disconnect the flexible tube, the absence of which greatly reduces the ‘drag’ on the $\varphi$ drive motor. Cryopumping remains efficient during several days of operation.

Temperature control is obtained with the aid of a heater resistor and a silicon-diode sensor attached to the second stage of the refrigerator cold head. The observed long-term stability is within 1 K. Without temperature control, temperatures slightly lower than 10 K were obtained but a variation of 0.5 K was observed between the two positions $\chi = 0$ and $\chi = -90^\circ$.

Using temperature control and at temperatures of 10 K and above, the short-term stability is approximately $\pm 0.2$ K. The measurement of the $\text{KH}_2\text{PO}_4$ phase transition at $T_c = 122.4$ K showed that the temperature gradient between the sensor and sample is less than 0.5 K.

The total transmission factor of the three aluminium (99.5% quality) environmental shields, measured using a neutron wavelength of 1.2 Å is 0.975 (3) when the Euler angle $\chi$ is zero. Owing to the relatively large diameter of the inner can ($D_3$) with respect to the usual sample dimensions, the neutron diffusion by the walls of the shields, has only a small effect on the background, provided that the collimation is kept to a reasonable minimum.

The system described has been made available for experiments for over six months. It is installed on D15 (Fig. 3), a four-circle diffractometer on the inclined thermal-neutron beam tube IH4 of the High-Flux Reactor of the Institut Laue-Langevin (ILL), Grenoble. One of the first experiments performed is reported by Filhol, Bravic, Lehner, Thomas (1980). In some experiments the normal beam geometry (lifting-counter technique) has also been used for data collection.

Drawings are available from Mr P. Simms at the ILL.

**References**


