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Strength of solid helium under high pressure

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Strength of solid helium under high pressure

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Abstract. Helium is commonly used as a pressure transmitting medium to render the sample pressure hydrostatic in high pressure experiments with diamond anvil cells. On solidification under pressure helium begins to develop strength that is characteristic of any solid. The estimation of the compressive strength of helium as a function of pressure is important as it helps in estimating the nonhydrostatic stresses that can develop in the sample even when it remains immersed in helium pressure transmitting medium. The x-ray diffraction data on polycrystalline samples obtained with the radial diffraction geometry are commonly used to determine the compressive strength of solids. This method fails in the case of helium because of its low scattering power. The nonhydrostatic stresses that develop in crystalline solids immersed in helium pressure transmitting medium were used to estimate the strength of helium. The diffraction data available in the literature were selected for this study. It was important for this study to consider the data from the experiments that were conducted ensuring that the samples did not come in contact with the anvils. The analysis of these data suggests that the strength of helium remains low (< 0.1 GPa) at pressures below 20 GPa and increases to ~ 2 GPa at 100 GPa.

1. Introduction

X-ray diffraction experiments using diamond anvil cells are routinely conducted to study a variety of pressure-induced effects in solids. For a meaningful interpretation of the high-pressure data and its comparison with the theoretical predictions, it is important to carry out the measurements under hydrostatic pressure. The pressure on the sample is rendered hydrostatic by containing the sample and a fluid pressure-transmitting medium in a metal gasket. The performance of a fluid as a pressure medium depends on its inability to support shear stresses. Ideally, the pressure medium should remain in fluid state and fully surround the sample. With increasing pressure, the fluids exhibit increase in viscosity and ultimately solidify at high enough pressures [1]. This leads to the appearance of nonhydrostatic stresses that increase with increasing pressure. Invariably, a pressure medium is much more compressible than the solid sample. This can result in the sample coming in contact with the anvils at pressures much below the pressure range where appreciable viscosity increase or solidification of the pressure medium occurs. The presence of the pressure medium in such cases becomes redundant. This problem has to be dealt with through careful assembly of the high-pressure cell. Helium, the best-known pressure medium, crystallizes under pressure [2, 3] but retains fluid-like flow property to much higher pressures than any other fluid. Eventually, it is expected to develop high mechanical stiffness to be able to support detectable nonhydrostatic stresses. These stresses are transmitted to the sample and the pressure on the sample begins to deviate from hydrostatic. It is therefore important to estimate the compressive strength of helium as a function of pressure. This will

define the lower limit of nonhydrostaticity of the sample pressure that will be present even when helium pressure medium is used.

Several investigators have used helium pressure-medium and followed as function of pressure the parameter like diffraction line-width or the quality of Brillouin scattering signals [4–8] to detect the onset of nonhydrostaticity. These studies suggest a hydrostatic -pressure limit that varies between 35 and 55 GPa. Such a wide range in the estimates, obviously, is due to the finer details of the loading of the sample and pressure medium in the metal gasket and the sensitivity to nonhydrostatic stresses of the parameter used for detection. In any case, these estimates are qualitative. The measurement of stresses has to be done through the measurement of strains these stresses produce. These strains (lattice strains) are derived from the shift of the diffraction-peak positions. Of course, appropriate elastic moduli are required to convert the measured strains to the stresses that produce them. Since the samples are in the form of polycrystalline aggregates, estimation of the appropriate elastic moduli is not straightforward. This necessitated the modelling of stresses in the sample compressed between the anvils [9, 10] and study of the effect of such stresses on the strains measured by x-ray diffraction [11–17]. The equations connecting the x-ray measured strains and stresses are commonly known as the lattice strain equations. These equations have been extensively used in deriving from the diffraction data the strength and elasticity of crystalline solids as a function of pressure. We present a brief account of this theory before discussing the strength of solid helium.

2. Lattice strain equations

The stress state of a solid sample under nonhydrostatic compression is axially symmetric about the load axis of the diamond anvil cell and is described by two equal stress components \mathbf{s}_{11} along the two orthogonal set of axes parallel to the anvil face and a component \mathbf{s}_{33} along the load axis [9, 10]. The off-diagonal terms in the stress tensor are assumed to be zero. The off-diagonal terms are often present in actual experiments. The effect of these terms on the lattice strain equations has been discussed in earlier publications [18, 19]. The difference between axial and radial stress components is termed uniaxial stress component or differential stress denoted by t . The maximum value of t is a measure of the compressive strength of the sample material at a pressure that equals the equivalent hydrostatic pressure (mean normal stress). The following relations can be written down

$$(\mathbf{s}_{33} - \mathbf{s}_{11}) = t \leq \mathbf{s}_Y \quad (2.1)$$

$$\mathbf{s}_P = (2\mathbf{s}_{11} + \mathbf{s}_{33})/3 \quad (2.2)$$

The term \mathbf{s}_Y denotes the yield strength of the solid sample at a confining pressure (equivalent hydrostatic pressure) \mathbf{s}_P . The d -spacing measured under such a stress state is given by [13–15]

$$d_m(hkl) = d_p(hkl)[1 + (1 - 3 \cos^2 \mathbf{y})Q(hkl)] \quad (2.3)$$

$$Q(hkl) = (t/6)\{\mathbf{a}[G_R^X(hkl)]^{-1} + (1 - \mathbf{a})(G_V)^{-1}\} \quad (2.4)$$

The subscripts m and P denote the quantities under nonhydrostatic (measured) and hydrostatic compression, respectively, and \mathbf{y} is the angle between the reflecting-plane normal and load axis of the diamond anvil cell. The term $G_R^X(hkl)$ is the diffraction shear modulus computed from the single-crystal elastic moduli under assumption of stress continuity across the boundaries separating the crystallites in the aggregate, G_V is the aggregate shear modulus computed under the assumption of strain continuity, and \mathbf{a} is a weight factor [13–15, 18]. Equations (2.1–2.4) are general and valid for all crystal systems. It is seen from equation (2.3) that, in presence of nonhydrostatic stresses, $d_m(hkl) > d_p(hkl)$ even though both are at same pressure \mathbf{s}_P . The differential stress is given by the following relation [15]

$$t = 6G\langle Q(hkl) \rangle \quad (2.5)$$

The term $\langle Q(hkl) \rangle$ represents the average of $Q(hkl)$ over all observed reflections. The parameter t is compressive and is taken to be positive by convention. The symbol G denotes the pressure dependent shear modulus. The measurement of t offers a unique method of determining the compressive strength of crystalline solids as a function of pressure. The use of equation (2.5) to determine t requires $d_m(hkl)$ versus \mathbf{y} data. Such data can be obtained with the radial diffraction geometry [17]. The strength of solid argon under pressure has been measured using radial diffraction technique, which gives the strength without any ambiguity. The strength of argon is small at low pressures but increases rapidly with pressure [20]. Helium too is expected to show similar behaviour. However, because of low scattering power for x-rays, recording of radial diffraction patterns of polycrystalline helium is challenging. Because of this difficulty we use the high-pressure x-ray diffraction data from polycrystalline solids recorded with helium pressure medium. The strength of helium is derived from the nonhydrostatic stresses it transmits to the sample. Such data have been recorded using the conventional diffraction geometry wherein the incident x-ray beam passes parallel to the load axis of the diamond anvil cell. In case of the cubic system, $a_m(hkl)$ versus $3(1-3\sin^2 \mathbf{q})\Gamma(hkl)$ plot (gamma plot) is a straight line given by [17],

$$a_m(hkl) \cong M_0 + M_1[3(1-3\sin^2 \mathbf{q})G(hkl)] \quad (2.6)$$

$$M_0 = a_p \{1 + (\mathbf{a}t/3)(1-3\sin^2 \mathbf{q})[(S_{11} - S_{12}) - (1 - \mathbf{a}^{-1})(2G_v)^{-1}]\} \quad (2.7)$$

$$M_1 = -a_p \mathbf{a}tS/3 \quad (2.8)$$

$$G(hkl) = (h^2k^2 + k^2l^2 + l^2h^2)/(h^2 + k^2 + l^2)^2 \quad (2.9)$$

$$S = (S_{11} - S_{12} - S_{44}/2) \quad (2.10)$$

$$t \cong -3M_1/[aM_0S] \quad (2.11)$$

Equation (2.11) suggests that the estimation of t from the diffraction data recorded using the conventional geometry depends on α . The value of α is not known *a priori*. In the early work in this field, α was assumed to lie between 0 and 1. However, it has been shown recently [18, 19] that this limit is valid only for solids with positive S , and $\alpha = 0.5$ is a good approximation in this case. For solids with negative S , α is greater than 1.

3. Strength of solid helium

For the present discussion we use the published x-ray diffraction data on niobium [21, 22], gold [8, 23, 24] and silver [25]. The data from these studies are chosen as the investigators ensured that the sample did not come in contact with anvils. Both S_p and t in such cases are transmitted to the sample by the helium pressure medium. These studies [21–25] were conducted to measure the equation of state under hydrostatic pressure. The data were analyzed in these studies to assess the hydrostaticity of pressure by estimating t in the polycrystalline samples using equation (2.11) and assuming $\mathbf{a} = 1$. In view of the recent studies [18, 19] it turns out that t derived with $\mathbf{a} = 1$ for gold was underestimated while for niobium overestimated [21–25]. We recalculated t for gold with $\mathbf{a} = 0.5$. The t -values for niobium published earlier [22] were recalculated using the \mathbf{a} -values as a function of pressure obtained in a recent work [19]. The data on niobium in helium pressure medium from another study [21] show abnormally large values and are not considered here. The recalculated t -values for gold and niobium are shown in figure 1. Also shown in figure 1 are the t -values of silver derived from the diffraction line-width analysis [25].

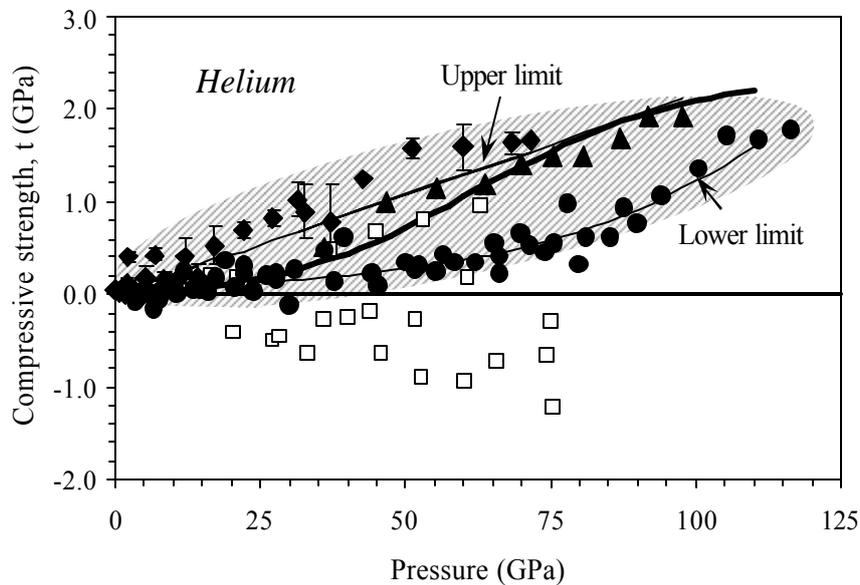


Figure 1. Recalculated values of t in niobium [22] – filled diamonds, gold [23, 24] – dots (ESRF data), and unfilled squares (Photon factory data). For details see [24]. Line-width analysis of silver data [25] – filled triangles. Suggested pressure dependence of strength of helium – thick line.

In case the samples are fully immersed in the helium pressure medium, the measured t should be independent of the sample material as it is t of the solid helium that is expected to be transmitted to the crystalline sample. Of course, implicit in this argument is the assumption that niobium, gold and silver have higher compressive strength than helium. Contrary to this expectation, the t - P data show considerable spread. Large spread is observed even among the data on the same material. For example, the t -values of gold exhibit a spread over negative to positive values. What is surprising is the occurrence of negative t . One set of data on niobium [22] also gave large negative t -values (not shown in figure 1). This situation does not arise when a solid sample is directly compressed between the anvils but is often observed in the experiments with helium pressure medium. Since $t = (\mathbf{s}_{33} - \mathbf{s}_{11})$, negative t would imply $\mathbf{s}_{11} > \mathbf{s}_{33}$. It is likely that highly compressible helium present in the cell as the pressure medium allows the metal gasket to collapse inward and leads to a situation where $\mathbf{s}_{11} > \mathbf{s}_{33}$. We term this as ‘abnormal compression’ of the gasket. The magnitude of the measured t in such cases depends on the deformation characteristic of the helium filled gasket. This possibly can explain rather large scatter observed in the t - P data shown in figure 1. If it were possible to fine-tune the deformation of the gasket, one could possibly achieve a condition with $\mathbf{s}_{33} = \mathbf{s}_{11}$, which would imply $t = 0$. From practical point of view this situation is of great interest as the unit cell parameters measured under this condition would be free from the systematic effect of nonhydrostatic stresses defined by equation (2.3). This, however, does not imply that pressure is truly hydrostatic. The shear stresses in the form of micro-stresses can still be present and the signatures of the resulting micro-strains could be seen in the diffraction line-broadening. The strength of the sample in such cases could still be determined from the line width analysis [26]. It may be pointed out that equation (2.11) could not be used to measure t in aluminium because it has small S but the line-width analysis could be used successfully [27]. It is important to point out that $M_1 = 0$ in the gamma plot occurs when either S or t

equals zero. As it is seen from equation (2.6), the systematic effect of nonhydrostatic stresses on the unit cell parameter still persists in case $S = 0$ but $t \neq 0$.

We exclude from the present discussion the negative t -values and consider only the data bound by the shaded region. The positive t -values show a lower limit predominantly determined by the t - P data (dots) of gold. The t - P data of silver and niobium mark the upper limit. Noting that t -values of gold also could be influenced by the abnormal compression of the gasket, it is tempting to conclude that the upper limit represents the strength of solid helium. However, we give credence to the data close to the lower limit and obtain a weighted average of the two limits. The t -values for niobium are influenced by the larger uncertainty in a for niobium in the low pressure range [19]. We assign arbitrarily higher weight (2:1) to the data lying close to the lower limit for pressures below 30 GPa. At higher pressure the weights are reversed. The averaged data are smoothened by fitting a function containing a three-parameter sigmoidal function and a linear term. The resulting curve is shown by the thick line in figure 1. To express the strength data as a fraction of the pressure-dependent shear modulus, we used the experimental data between 13 and 32 GPa [28]. We fitted a two-parameter logarithmic equation to the measured data and used this equation to compute shear moduli at required pressures. The commonly used finite strain equation [29] could not be used because only six data points were available for this fit. The strength of solid helium remains small ($< 0.005G$) at pressures below 20 GPa and increases to $0.03G$ at 100 GPa. These data should be considered only rough estimates because of the uncertainties in the extrapolation of the shear modulus data over a large pressure range.

4. Discussion

The analysis of diffraction data recorded under nonhydrostatic compression gives the uniaxial stress component t . For t to be equal to the yield strength it is important that the maximum value of t , which the sample material can support, should develop in the experiments. In most cases, this condition is achieved by compressing the solid sample directly between the anvils without any pressure medium. In such cases, the gasket begins to deform upon compression by outward flow. Such deformation is essential for the development of maximum t in the sample. The situation is different when the gasket contains highly compressible material like helium. The deformation of the gasket may begin with inward flow of the gasket. This gives rise to the t -values that are lower than the maximum and, in the extreme case, can result in negative t . There is no way of ensuring the development of maximum t in these experiments. For this reason, the strength of helium obtained here may have been underestimated. The t -values derived from the diffraction data recorded with the conventional geometry is influenced by the choice of a . Uncertainty in t arising from the uncertainty in a could be as high as $\pm 20\%$. The uncertainty in t from this source could be eliminated if the radial diffraction data become available but the effect of abnormal compression of the gasket will continue to persist.

Tempere and Silvera [30] recently analyzed the pressure distribution inside the cell containing solid sample (pressure marker) and helium pressure medium using finite element analysis. These investigators show that the pressures of the sample and pressure medium differ. This difference is a function of the elastic properties of the sample and pressure medium. Further, pressure gradients exist in the pressure medium in the vicinity of the sample. In the present study, the pressure of the medium is inferred from the pressure of solid sample. The difference between the two pressures is expected to be only a few gigapascal at the highest pressure. The second factor does not affect the diffraction line-shift and, therefore, will not affect t determined from the gamma plots. However, the strength of silver inferred from the line-width analysis may be in error. In view of the overall uncertainty in estimating the strength of solid helium, the influence of both the factors is not significant.

The strength of argon, another inert gas, was determined as a function of pressure from the radial diffraction data in an earlier study [20]. The strengths of argon and helium are compared in figure 2. The strength of argon is nearly zero at low pressure and increases steeply as the pressure is increased. The strength, however, is unlikely to continue increasing at this rate and is bound to exhibit a gradually decreasing slope such that the strength asymptotically approaches infinity at infinite pressure. In comparison, helium also has near-zero strength at low pressures and increases with

increasing pressure but not as steeply as for argon. At 50 GPa, the strength of argon is nearly seven times the strength of helium.

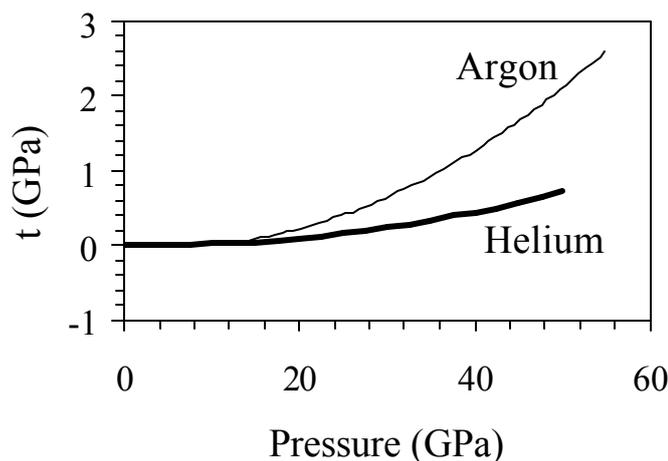


Figure 2. A comparison of the compressive strengths of helium and argon. Thick line – helium (this study). Thin line – argon. The strength of argon was determined from the radial x-ray diffraction data [20].

5. References

- [1] Angel R J, Bujak M, Zhao J, Diego Gatta G D and Jacobsen S D 2007 *J. Appl. Cryst.* **40** 26–32
- [2] Loubeyre P, LeToullec R, Pinceaux J P, Mao H K, Hu J and Hemley R J 1993 *Phys. Rev. Lett.* **71** 2272–5
- [3] Mao H K, Hemley R J, Jephcoat A P, Finger L W, Zha C S and Bassett W A 1988 *Phys. Rev. Lett.* **60** 2649–52
- [4] Bell P M and Mao H K 1981 *Carnegie Inst. Washington Publ.* **80** 404
- [5] Alexandrov L V, Gonchrov A F, Zosman A N, and Stishov S M 1987 *Sov. Phys. JET* **66** 384
- [6] Zhang L, Ahsbahs H, and Kuteglu A 1998 *Phys. Chem. Miner.* **25** 301
- [7] Zha C S, Mao H K, and Hemley R J 2000 *Proc. Natl. Acad. Sci. U.S.A.* **97** 13494–9
- [8] Takemura K 2001 *J. Appl. Phys.* **89** 662–8
- [9] Singh A K and Kennedy G C 1974 *J. Appl. Phys.* **45** 4686–91
- [10] Ruoff A L 1975 *J. Appl. Phys.* **46** 1389–92
- [11] Singh A K 1993 *J. Appl. Phys.* **73** 4278–86: Erratum 1993 *J. Appl. Phys.* **74** 5920
- [12] Uchida T, Funamori N and Yagi T 1996 *J. Appl. Phys.* **80** 739–6
- [13] Singh A K, Balasingh C, Mao H-k, Shu J and Hemley R J 1998 *Rev. High Pressure Sci. Technol.* **7** 205–10
- [14] Singh A K, Mao H-k, Shu J and Hemley R J 1998 *Phys. Rev. Lett.* **80** 2157–60
- [15] Singh A K, Balasingh C, Mao H-k, Hemley R J and Shu J 1998 *J. Appl. Phys.* **83** 7567–75
- [16] Matthies S, Priesmeyer G and Daymond M R 2001 *J. Appl. Cryst.* **34** 585–601
- [17] Singh A K 2004 *J. Phys. Chem. Solids* **65** 1589–96
- [18] Singh A K 2009 *J. Appl. Phys.* **106** 043514
- [19] Singh A K and Liermann H P 2011 *J. Appl. Phys.* **109** 113539
- [20] Mao H-k, Badro J, Shu J, Hemley R J and Singh A K 2006 *J. Phys.: Condens. Matter* **18** S963–8
- [21] Singh A K and Takemura K 2001 *J. Appl. Phys.* **90** 3269–75: Erratum Singh A K and Takemura K 2002 *J. Appl. Phys.* **91** 5507
- [22] Takemura K and Singh A K 2006 *Phys. Rev. B* **73** 224119
- [23] Dewaele A, Loubeyre P and Mezouar M 2004 *Phys. Rev. B* **70** 094112
- [24] Takemura K and Dewaele A 2008 *Phys. Rev. B* **78** 104119

- [25] Dewaele A and Loubeyre P 2007 *High Press. Res.* **27** 419–29
- [26] Singh A K, Liermann H-P and Saxena S K 2004 *Solid State Commun.* **132** 795–8
- [27] Singh A K, Liermann H-P, Akahama Y and Kawamura H 2007 *J. Appl. Phys.* **101** 123526
- [28] Zha C-S, Mao H-k and Hemley R J 2004 *Phys. Rev. B* **70** 174107
- [29] Birch, F 1978 *J. Geophys. Res.* **83**, No. B3 1257–68
- [30] Tempere J and Silvera I F 2011 *J. Appl. Phys.* (under publ.)