

# MgO INTERFEROMETRIC SENSOR FOR HIGH PRESSURE MEASUREMENT UNDER HIGH TEMPERATURES

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A high pressure sensor, based on the shift of the interference peaks of a MgO etalon was developed. Measurement of the pressure dependence of the refractive index of MgO up to 10 GPa was performed, being well described by the Lorentz–Lorenz equation with a constant Mueller parameter of 1.28. These results, together with other well-known properties of MgO, enable the proposition of an equation for the shift of the interference peaks with pressure and temperature. This equation can be used for highly accurate pressure measurements, even at high temperatures. The application of the method to different types of pressure chambers is discussed.

## 1. Introduction

Techniques for high pressure measurements under temperature are of great importance in phase diagrams studies, particularly those related to superhard materials production. The ruby fluorescence technique has been extensively used for pressure measurements in diamond anvil cells. However, it is limited, in practical use, to temperatures lower than 600 K. Recently an alternative method for pressure measurement was demonstrated, based on optical interferometry, which can be used at high temperatures.<sup>1)</sup> In that work, pressure was determined by the shift in wavelength ( $\Delta\lambda$ ) in the interference peaks of a small etalon placed inside a diamond anvil cell (DAC), given by:

$$\begin{aligned}\Delta\lambda &= \lambda(0) \left[ \frac{l(P, T)}{l(0, T)} \frac{n(P, T)}{n(0, T)} - 1 \right] \\ &= \lambda(0)G(P, T)\end{aligned}\quad (1)$$

where  $l$  and  $n$  are the sample thickness and refractive index, respectively, and  $G(P, T)$  is the gauge function, depending on pressure and temperature.

The precision of the method is then related to the precision in the determination of the pressure ( $P$ ) and temperature ( $T$ ) dependences of  $n$  and  $l$ ,

since the peak widths are only dependent on the surface flatness and parallelism, and on the coating reflectivity—factors that are not strongly affected by pressure or temperature.

The dependence of volume on  $P$  and  $T$  are fairly accurately known for a number of crystals, including MgO.<sup>2),3)</sup> However, the pressure dependence of the MgO refractive index is only known up to 0.7 GPa, from the work of Vedam and Schmidt.<sup>4)</sup>

In this work, a small etalon of MgO was subjected to high hydrostatic pressures, up to 10 GPa, in the DAC. Through the shift of the interference peaks, we measured the pressure dependence of  $n$  for MgO, in order to obtain the calibration function  $G(P, T)$ .

## 2. Theoretical approach

A common approach to the refractive index in solids is to consider the individual ions as polarizable entities, with polarizabilities  $\alpha$ . The total electric field acting upon one ion is, then, the sum of the external field and the field produced by the neighbor dipoles, accordingly the Clausius–Mossotti (Lorentz–Lorenz) classical model.

Despite criticism of this approach, based on the inadequacy of the Lorentz–Lorenz (LL) classical model in comparison with a more fundamental

quantum mechanical approach, as pointed out by Pantelides,<sup>5)</sup> it is a simple and attractive picture, that well describes the experimental results for several insulators. The LL relation is given by:

$$\frac{n^2 - 1}{n^2 + 2} = \frac{4\pi}{3} N\alpha, \quad (2)$$

where  $N$  is the ion density.

Mueller<sup>6)</sup> considered this relation valid, provided that  $\alpha$  was volume dependent, and introduced the strain polarizability constant, or Mueller parameter,  $\Lambda_0$ , to account for this volume dependence:

$$\Lambda_0 = \left( \frac{\partial \ln \alpha}{\partial \ln V} \right)_T. \quad (3)$$

For MgO, it is known<sup>7)</sup> that  $\alpha$  is approximately independent of  $T$  explicitly, so the temperature change on  $n$  would be generated only indirectly due to the change of  $V$ .

On the other hand, the dependence of  $l$  on  $P$  and  $T$  may be given by the following equation of state, recently proposed by Guillermet *et al.*<sup>8)</sup>:

$$V(T, P) = V(0, 0) [1 + B'_0 P / B_0(T)]^{-1/B'_0} e^{\beta_0 T + \beta_1 T^2/2}, \quad (4)$$

where the factor between square brackets is the well-known Murnaghan equation of state,  $B_0(T)$  being the isothermal bulk modulus and  $B'_0$  being a constant. The exponential factor is obtained by integration of the assumed linear temperature variation of the thermal expansivity,  $\beta(0, T) = \beta_0 + \beta_1 T$ . We considered the following dependence of  $B_0$  on  $T$ :

$$B_0(T) = B_{00} + B_1 T + B_2 T^2, \quad (5)$$

$B_{00}$ ,  $B_1$  and  $B_2$  being constants.

### 3. Experimental procedure

The MgO etalon was manually polished down to a thickness of 40  $\mu\text{m}$ , with parallel faces covered by partially reflecting metallic films. It was inserted, together with a ruby calibrant, into the

gasketed chamber of the DAC, resting horizontally over the anvil surface. Nearly parallel light from a high-pressure xenon lamp was used to perpendicularly illuminate the etalon, yielding a channeling spectrum for the transmitted light, with interference peaks. A special collimating system was used to improve the peak-to-background ratio. This basically consists of focusing the image of a pinhole source entirely onto the etalon surface, reducing the scattering of the light by diamond faces, and avoiding the incidence of a direct beam in the spectrometer. Figure 1 shows a typical interference pattern obtained with the etalon inside the DAC.

Following one peak in a wide pressure range, up to 10 GPa, and using Eq. (1) together with the known pressure dependence of  $V$  for MgO, described by Eq. (4), we obtained the pressure dependence of the MgO refractive index at room temperature. The small dispersion of  $n$  in the covered spectral range was also considered.

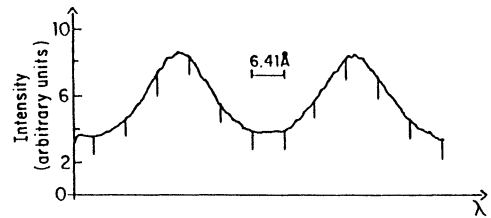


Fig. 1. Typical interference pattern of the MgO etalon inside the DAC, under high pressure. The wavelengths are about 7000 Å.

### 4. Results and discussions

With the experimental results for  $n(P)$  and the LL relation we obtained the volume dependence of  $\alpha$  for MgO at room temperature, which was practically linear. We found, through a least square fit of the data, a constant Mueller parameter of 1.28, in agreement with Vedam and Schmidt,<sup>4)</sup> despite the fact that they found an increasing behavior of  $\Lambda_0$  with pressure up to 0.7 GPa, following a different procedure.

Using these results and considering the LL approach, together with Eq. (4) for  $V(T, P)$ , it is possible to write the following calibration function:

$$G(P, T) = \frac{A'x^{1/3}[(A + x^{\Lambda_0-1})/(A - x^{\Lambda_0-1}/2)]^{1/2} - 1}{1 - x^{1/3}\partial \ln n / \partial \ln \lambda}, \quad (6)$$

where  $A=(n_0^2+2)/(n_0^2-1)$ ,  $A'=\sqrt{(A-0.5)/(A+1)}$ ,  $n_0=n(V_0, T_0)=1.736$ , and:

$$x = V(P, T) / V(0, T_0) = 0.991[1 + B_0'P/B_0(T)]^{-1/B_0'} \cdot \exp(\beta_0 T + \beta_1 T^2/2), \quad (7)$$

$T_0$  being the room temperature. The denominator in  $G(P, T)$  accounts for the small dispersion in  $n$  due to the spectral range covered, 6880 Å to 7010 Å, that is approximately  $\partial n / \partial \lambda = -5 \times 10^{-6} \text{ Å}^{-1}$ . For MgO, the parameters of  $G(P, T)$  are:  $B_0'=4.54$ ,<sup>9)</sup>  $\beta_0=2.768 \times 10^{-6} \text{ K}^{-1}$ ,<sup>2)</sup>  $\beta_1=1.968 \times 10^{-8} \text{ K}^{-2}$ ,<sup>2)</sup>  $B_{00}=170 \text{ GPa}$ ,<sup>10)</sup>  $B_1=-0.0247 \text{ GPa K}^{-1}$ ,<sup>10)</sup> and  $B_2=-2.706 \times 10^{-6} \text{ GPa K}^{-2}$ ,<sup>10)</sup> pressure being given in GPa and temperature in K.

Figure 2 shows the experimental measurements for the shift of the peaks with pressure, together with the calibration function, Eq. (6), where the continuous line represents  $G(P, T)$  for room temperature, and the dotted line,  $G(P, T)$  for 500 K. As can be seen, the agreement between the experimental results and the proposed function is excellent, at least at room temperature. However, we believe it will also remain good for high

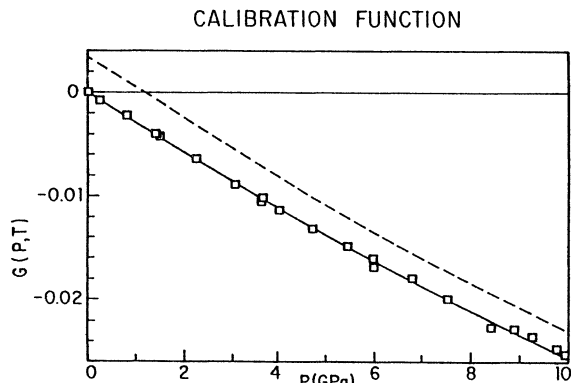


Fig. 2. Experimental results for the shift of the interference peaks with pressure at room temperature, together with the calibration function,  $G(P, T)$  for room temperature (continuous line) and for 500 K (dotted line).

temperatures, since the calibration function, Eq. (6), contains a well-studied equation of state,  $V(T, P)$ , for MgO, and the volume dependence of  $\alpha$  was experimentally determined through the LL relation in this work. Despite the fact that polarizability generally changes explicitly not only with volume but also with temperature, it is known that, for MgO, there is practically no explicit dependence on temperature.<sup>7)</sup> Nevertheless, an eventual slight correction in the high temperature range could be made by fine adjustments in some of the parameters Eq. (6).

We observed a pressure shift of  $-20 \text{ Å/GPa}$  and a linewidth of about  $17 \text{ Å}$  for the MgO etalon, while for ruby, the pressure shift of the fluorescence peaks is  $3.6 \text{ Å/GPa}$ , and the linewidth obtained was approximately  $7 \text{ Å}$ . Therefore, the accuracy in pressure measurements using this interferometric procedure, was potentially more than twice as high as that of the ruby method, and can be further improved through better sample preparation, being in principle unaffected by temperature. It is interesting to remark that for the ruby method, there is a strong broadening of the fluorescence peaks with increasing temperature, which limits its applicability to temperatures below 600 K.

The method is applicable to high-pressure chambers having an optical window and a hydrostatic environment for the sensor. For example, in a belt apparatus, the lower piston can be fitted with a diamond window, followed by a conic exiting hole, as described in Ref.<sup>11)</sup>. A hydrostatic capsule must surround the calibrant, and the interference peaks are than obtained by reflection. Such implementation is now being made and will soon be reported.

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