

High temperature dilatation measurements by *in situ* laser interferometry under high magnetic field

S. Rivoirard,^{a)} T. Garcin, E. Beaugnon, and F. Gaucherand
CNRS-CRETA, BP166, 38042 Grenoble Cedex 9, France

(Received 18 May 2009; accepted 1 September 2009; published online 1 October 2009)

This article contains a description of a laser interferometer coupled with a furnace for *in situ* dilatation measurements under high magnetic field. The apparatus fits an 18 T superconducting magnet with a 32 mm diameter room temperature bore. This optical method was chosen for dilatation measurements because it is not perturbated by the magnetic field. The measured sample can be heated up to 1500 K under a controlled atmosphere, and heating and cooling rates can be varied within a range of 10 K/s. The resolution is below 50 nm. As an example of using *in situ* dilatation measurements to follow phase transformations, the increase in the ferrite/austenite transformation temperature in pure iron in a magnetic field up to 16 T was experimentally evidenced. © 2009 American Institute of Physics. [doi:10.1063/1.3234344]

I. INTRODUCTION

Magnetic field processing is a promising new tool for the structural and functional control of materials. A significant potential exists for tailoring microstructures of materials by using a magnetic field during the fabrication process. Recently, remarkable achievements of cryocooled superconducting magnets enable the easier use of high magnetic fields in large-scale room temperature spaces. With this new development, magnetic field processing has become more attractive. There has been a lot of work to control the morphology of materials by using a magnetic field. As an example, in ferromagnetic alloys, structural changes may result from the influence of the magnetic field on thermodynamic equilibria. Phase equilibria can be shifted by the application of a magnetic field, as a result of the modification of the partial molar Gibbs free energy of each species.^{1–4} In this case, the magnetic field acts toward the stabilization of the phase with the highest magnetization and hence transformation processes and kinetics can be modified.^{5,6}

Recently, a need for new devices to monitor high temperature phase transformations under magnetic field has emerged. Beside post-treatment metallographic analyses of the microstructure resulting from high magnetic field processing,^{7,8} it is necessary to design experiments for the *in situ*, real time, control of the phases transformations. Several techniques have already been proposed, using magnetization,^{9–11} electrical resistivity,¹² and thermal analysis.¹³ High temperature magnetization measurement is a powerful tool to probe the phase transformations such as melting, solidification and solid state transformation. Because the measurement is based on the force exerted on the magnetized sample placed in a field gradient, additional phenomena due to the magnetic torque can be observed and limited field intensities are applied to the sample in this position. Electrical resistivity has been recently used to probe

the austenite /ferrite transformation of pure Fe in high magnetic fields up to 10 T.¹² The austenite/ferrite transformation is seen by a small change in the resistivity and the transformation temperature is clearly determined although the signal remains noisy.

Thermal analyses are also a simple and efficient way to evidence endothermic or exothermic transformations.¹³ This technique requires a medium cooling or heating rate and the onset, rather than the full transition is monitored.

Dilatometry is a powerful and common technique used for the study of phase transformations in materials and more particularly, for the study of the transformation behavior of steels during continuous heating, cooling and isothermal holding.¹⁴ A dilatometer includes either a furnace or a cryostat depending on the temperature range required for measurements. On the basis of the measured sample length changes with temperature, which are directly correlated with changes in the specific volume, solid state phase transformations can be investigated using this technique. In the magnetic field vicinity, an optical technique for dilatation measurement seems to be a natural choice. To our knowledge, laser dilatometry has never been used in high magnetic field experiments. Consequently, we developed a high magnetic field dilatometer, ranging from room temperature up to 1500 K, using a high resolution Michelson laser interferometer. In this paper, the apparatus is described and dilatometry is used to monitor the austenite to ferrite transformation in pure iron.

II. APPARATUS

The experimental setup, presented in Figs. 1 and 2, consists of a high temperature resistive furnace inserted into a superconducting magnet and a laser interferometer measuring the sample dilatation. The magnet (Oxford Instrument) can reach 16.5 T at 4.2 K, when operating at 2 K by pumping the helium bath, the maximum field is 18.5 T. The available experimental space inside the magnet is a vertical cylindrical room temperature bore with a diameter of 32 mm. The ver-

^{a)}Electronic mail: sophie.rivoirard@grenoble.cnrs.fr.

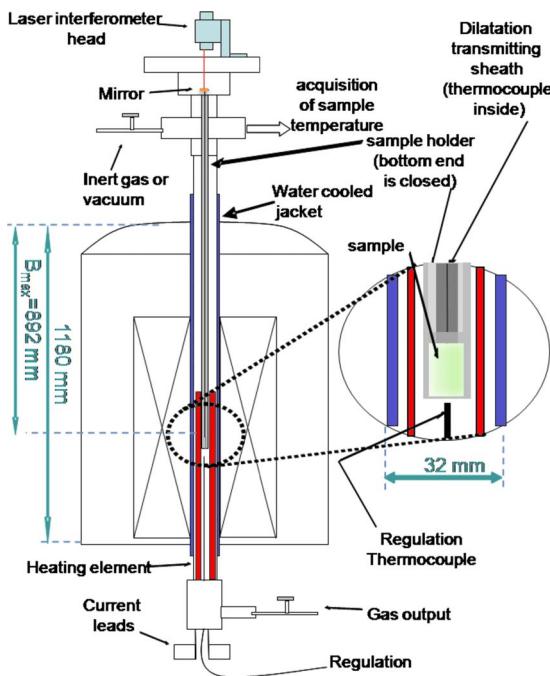


FIG. 1. (Color online) Experimental measurement setup in the superconducting magnet: the furnace consists of a water cooled jacket of a heating element (inserted from the bottom) and is closed on top by the sample holder. The dilatation measurement is realized through the beam reflection on the mirror screwed on top of the transmitting sheath in contact with the upper surface of the sample.

tical size of the bore is 118 cm, and the maximum field is located at 89.2cm below the magnet top flange. To protect the magnet from the furnace heating, a water cooled jacket with a 26 mm inner diameter is inserted along the whole vertical bore of the magnet.

The high resolution Michelson laser interferometer is a HC 500 micromodel (by Sageis-CSO, Grenoble, France). The laser diode, the optical parts and the detectors are inte-

grated in a pen-size head (about 10 cm in height and 1cm in diameter) placed above the magnet entrance. The laser beam has a wavelength of $\lambda=780$ nm and a diameter of 1 mm. The measuring beam (one of the two branches of a Michelson interferometer) crosses the Pyrex furnace windows and is reflected by a spherical mirror connected to the sample through an alumina sheath. The dilatation of the sample vertically displaces this sheath together with the mirror screwed on top of it. The reflected measuring beam interferes with the reference beam located in the interferometer head itself and the relative dimensional change in the sample is measured through the displacement of the interference fringes between the reflected beam and the reference beam. The reference beam never leaves the interferometer head. As a result, the changing interference pattern comprises both the sample dilatation itself and other contributions (sample holder, alumina connecting sheath, furnace's frame). Reference experiments (without sample and with standard probes such as silicon or alumina replicas) are regularly performed, to measure the apparatus contribution to the dilatation measurement. These reference measurements enable to obtain the background signal, by subtracting the standard probe dilatation coefficient expected from literature data. This background contribution is found to have a linear variation with temperature, which amounts to around 150 μm over the full temperature range of the instrument. This background contribution is then further removed from the measured signal to obtain the dilatation of the sample.

The alumina sheath enables not only the deformation of the sample to be transmitted but also the temperature of the sample to be measured. A Pt/Pt-Rh thermocouple at the bottom end of this sheath is in close contact with the sample to measure its temperature (accuracy is ± 1 K).

The interferometer control unit provides several outputs: a high frequency digital connection to a computer allows the continuous measurement of the mirror displacement and two analog outputs provide the cosine and the sine of the phase delay between the measuring and reference beams. These two outputs are connected to an oscilloscope to conveniently monitor the signal strength and stability. From the signal width on the scope, the resolution in our operating conditions is estimated to be below 50 nm. The final accuracy depends on the whole furnace dilatation compensation.

The furnace consists of a spiraling, hollow, silicon carbide heating element with an inner diameter of 9.5 mm. It is inserted into the magnet from the bottom of the magnet bore and fixed to the water jacket. A dc power supply is used rather than an ac power supply to avoid strong vibrations due to the coupling between the magnetic field and the current. In the few millimeters between the cold jacket and the heater, a 2 mm thickness alumina wool sheet provides sufficient thermal insulation. The upper part of the jacket is closed by a Pyrex window and the lower part is sealed so that the furnace can operate under a secondary vacuum or controlled atmosphere. The temperature is controlled through a Eurotherm controller and can reach 1500 K, with heating and cooling rates in the range of 10 K/s.

The sample holder consists of a brass tube connected to the top of the jacket and of an alumina tube, which is sus-

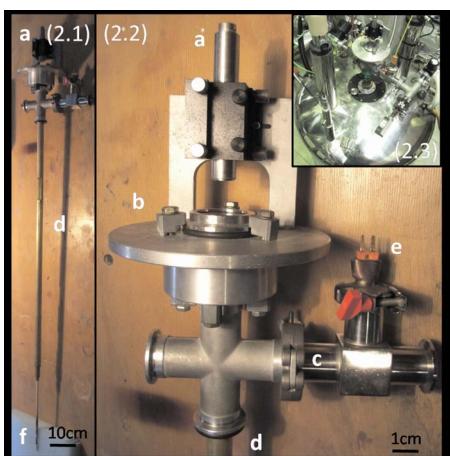


FIG. 2. (Color online) Photographs of (2.1) full sample holder and guiding system, (2.2) upper part of the dilatometer apparatus, and (2.3) top view of the insertion of (2.1) in the superconducting magnet. (a) Laser head mounted into the xyz moving stage, (b) airtight Pyrex window, (c) vacuum-gas connections, (d) guiding tube, (e) thermocouple output, and (f) alumina sample holder.

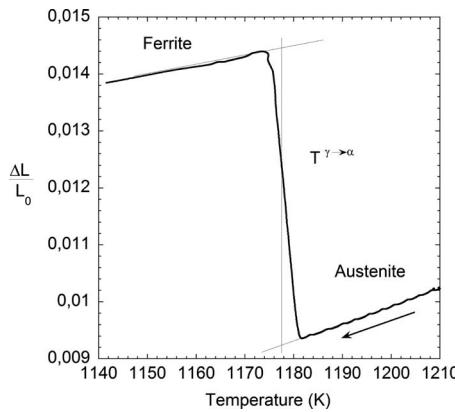


FIG. 3. Change in the sample length relative to the initial (room temperature) length as a function of temperature recorded during the austenite to ferrite transformation without applied magnetic field.

pended below, with its lower part inside the hollow heating element. The sample, 4 mm in diameter and 10 mm in length, stands on the bottom closed end of the alumina tube and is positioned in the homogeneous temperature region of the heating element. This position coincides with the homogeneous magnetic field zone, so that the bottom of the sample never changes position during measurements. Special care must be taken to set up the sample in the position where the vertical magnetic forces balance near zero. If the position is too low, the sample is lifted and a contribution is added to the dilatation signal. If the position is too high, the magnetic force pushes the sample toward the bottom of the sample holder and induces an additional elastic deformation contribution. By recording the dilatation signal at room temperature while raising the field, any small variation of the sample position due to the magnetic force shows up immediately. This enables to adjust the sample equilibrium position before starting the experiment. To prevent from the magnetic torque, the 10 mm sample length is vertical and parallel to the magnetic field. In this case, no torque is induced by the sample shape anisotropy (the ratio of length over diameter is much higher than 1). As a second precaution, the inner diameter of the alumina tube (sample holder) is adjusted to the sample diameter. Thus, neither magnetic forces nor torques are a cause of perturbation during measurements (within experimental resolution). On top of the sample holder, an evacuation pipe is connected to a secondary vacuum pump and an electrovalve enables the furnace chamber to be filled with purified gas: He, Ar, Ar/H₂, or N₂ can be chosen as the experimental environment. A Pirani–Penning vacuum gauge, together with a manometer is also connected to the furnace chamber through this upper part. On the sample holder and above the vacuum/gas connections, a stand with a xyz fine-motion stage is fixed to maintain the laser head in the vertical position. This set stands 25 cm above the entrance of the magnet bore. At this position, no perturbation due to the magnetic field has been observed in the signal delivered by the laser head detectors. The xyz fine-motion stage is used to set the operating point and to adjust the alignment of the laser beam.

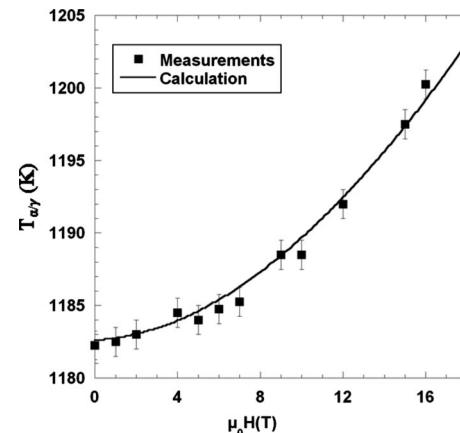


FIG. 4. Variations in the austenite/ferrite equilibrium temperature, $T_{\alpha/\gamma}$, as a function of applied magnetic field.

III. HIGH FIELD DILATOMETRY RESULTS

Figure 3 shows the change in the sample length (ΔL) relative to the initial length at room temperature L_0 . In this figure, the contraction of a high purity iron sample is observed on cooling (1 K/min). The γ/α transition shows up as a sharp expansion of the sample. The measured volume change during the transition is about 1.5%, in good agreement with the calculated density ratio of the two phases in this temperature range. Expansion coefficients of $15 \cdot 10^{-6} \text{ K}^{-1}$ for ferrite (α) and $25 \cdot 10^{-6} \text{ K}^{-1}$ for austenite (γ) are deduced from this measurement.

The effect of magnetic field on pure iron was tested for several values between 0 and 16 T. The field dependence of the equilibrium transformation temperature $T_{\alpha/\gamma}$ in iron is shown in Fig. 4. The transformation temperatures are noted $T_{\alpha \rightarrow \gamma}$ on heating and $T_{\gamma \rightarrow \alpha}$ on cooling. $T_{\alpha \rightarrow \gamma}$ (respectively, $T_{\gamma \rightarrow \alpha}$) is defined as the temperature at which the linear thermal expansion, graphically represented by the $\Delta L/L_0 = f(T)$ function in Fig. 3, deviates from linearity. Location of the point at which the deviation occurs is obtained by extrapolating the linear portions of the thermal expansion curve. The equilibrium temperature is then obtained by $T_{\alpha \rightarrow \gamma} = (T_{\alpha \rightarrow \gamma} + T_{\gamma \rightarrow \alpha})/2$. It is plotted as a function of the magnetic field intensity in Fig. 4. The variation relative to the equilibrium temperature in the absence of magnetic field, amounts to 17 K in pure Fe, in a magnetic field of 16 T. This increase is proportional to the square of the magnetic field. The ferrite is stabilized at the expense of austenite. This result constitutes strong experimental evidence of the predicted calculations¹ based on the Weiss molecular field model (solid line in Fig. 4) and validates the use of high field laser dilatometry for such investigations.

To conclude, a laser interferometer has been developed under a magnetic field of 18 T to allow real time dilatation measurements up to 1500 K with sensitivity below 100 nm. Experimental investigations of the magnetic field effects on high temperature phase transformations in a wide range of magnetic alloys are expected from this new device.

¹H. Guo and M. Enomoto, Mater. Trans., JIM **41**, 911 (2000).

²J. K. Choi, H. Ohtsuka, Y. Xu, and W. Y. Choo, Scr. Mater. **43**, 221 (2000).

- ³H. D. Joo, S. U. Kim, N. S. Shim, and Y. M. Koo, *Mater. Lett.* **43**, 225 (2000).
- ⁴M. C. Gao, T. A. Bennet, A. D. Rollet, and D. E. Laughlin, *J. Phys. D* **39**, 2890 (2006).
- ⁵G. M. Ludtka, R. A. Jaramillo, R. A. Kisner, D. M. Nicholson, J. B. Wilgen, G. Mackiewicz-Ludtka, and P. N. Kalu, *Scr. Mater.* **51**, 171 (2004).
- ⁶Y. Zhang, C. He, X. Zhao, L. Zuo, and C. Esling, *J. Magn. Magn. Mater.* **294**, 267 (2005).
- ⁷Y. Zhang, C. Esling, M. Calcagnotto, M. L. Gong, X. Zhao, and L. Zuo, *J. Phys. D* **40**, 6501 (2007).
- ⁸S. Rivoirard, F. Gaucherand, E. Beaugnon, O. Bouaziz, and E. Pinto Da Costa, *Rev. Metall./Cah. Inf. Tech.* **5**, 393 (2005).
- ⁹F. Gaucherand and E. Beaugnon, *Physica B* **294-295**, 96 (2001).
- ¹⁰S. Rivoirard, F. Gaucherand, O. Bouaziz, E. Pinto da Costa, and E. Beaugnon, *ISIJ Int.* **46**, 1274 (2006).
- ¹¹T. Ono, K. Sassa, K. Iwai, H. Ohtsuka, and S. Asai, *ISIJ Int.* **47**, 608 (2007).
- ¹²H. Miyauchi, T. Fukuda, and T. Kakeshita, *CAMP-ISIJ* **17**, 1227 (2004).
- ¹³X. Hao and H. Ohtsuka, *Mater. Trans.* **45**, 2622 (2004).
- ¹⁴C. Garcia de Andres, F. G. Caballero, C. Capdevila, and L. F. Alvarez, *Mater. Charact.* **48**, 101 (2002).