Non-contact ultrasonic measurements of the elastic constants of magnetic materials

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Abstract. Ultrasonic testing using contacting transducers such as quartz or PZT is well established. However, standard measurement techniques used require physical contact of the sample and ultrasonic transducer and some sort of couplant between the two. With this configuration there is a possibility of damaging the sample, transducer or bond during testing, thermal cycling, or removal of the transducer. We present results taken using recent advances in non-contact methods of ultrasound generation and detection using electromagnetic acoustic transducers (EMATs), which offer some significant benefits over contact ultrasonic techniques. Circumventing the need for couplant removes the possibility of contaminating the system, which is an issue for some material property measurements, and allows easier measurements over a wider range of temperatures. An automated data analysis system has been developed which allows the velocity of sound in the sample, and hence the elastic constants, to be determined to a high accuracy. This technique is illustrated using measurements of the alloy Gd$_{64}$Sc$_{36}$.

1. Introduction
Materials testing using contact transducers for generation and detection of ultrasonic waves is well used in areas ranging from medical physics, to non-destructive testing (NDT), through to fundamental physics experiments [1, 2, 3, 4, 5, 6]. Many of the future improvements in ultrasonic measurements are likely to be through developments in non-contact methods of generation and detection, as these offer significant benefits over contact ultrasonic techniques [2].

Standard ultrasonic measurements of single crystals use contact transducers, such as quartz or LiNbO$_3$, which require contact with the sample and couplant for transmission of the waves [6]. Repeated thermal cycling can affect this couplant, causing damage, dramatically reducing the coupling efficiency of the transducer to the sample, and ultimately leading to a complete loss of signal. Recent developments in NDT [2] have ensured that electromagnetic acoustic transducers (EMATs) are now a practical alternative to contact techniques.

Figure 1(b) shows two EMAT designs used for non-contact ultrasonic measurements, with lift-offs from the sample possible of up to several mm. The first is a typical EMAT used for NDT, which consists of a coil and a permanent magnet, with a current pulsed through the coil for generation of ultrasound [2]. These operate on electrically conducting samples (via the Lorentz force mechanism) and/or certain magnetic samples (via magnetoelastic mechanisms). For the Lorentz force mechanism the electrons in the mirror current in the sample experience a force and in turn ‘drag’ the atoms, with each current pulse generating an ultrasound pulse [2]. In magnetic materials, ultrasound is induced by the varying magnetic field from the current pulse.
Through careful consideration of the coil and magnet configuration various ultrasonic waves can be generated [7]. Detection is via a similar mechanism [2]. During measurements of the properties of a crystal the effects of temperature and magnetic field may be investigated [8, 9, 10]. In this case, the magnetic field for these experiments can be used as part of the EMAT in the place of the permanent magnet.

![Figure 1](image.png)

**Figure 1.** (a) shows a typical ultrasound signal from a single crystal, showing ultrasonic echoes with separation $\Delta t$. (b) shows EMATs developed for NDT (left) and single crystal measurements (right, after [8]).

EMATs have been used for some recent measurements of single crystals [8, 9, 11]; however, practically there are still significant issues. EMATs are, by their very nature, sensitive to electrical noise, and their signal to noise ratio when compared with standard techniques can be poor if the environment is electrically noisy. Filtering and lock-in techniques can be used to remove some of this noise, but must be used carefully to ensure that none of the information included within the signals is lost. However, the advantage of the no couplant requirement still makes EMATs an attractive prospect. The efficiency of non-contact ultrasound generation is significantly lower than that obtained when using piezoelectric transducers, and the signal to noise ratio can be an issue even without electrical noise. However, EMATs are highly sensitive to magnetic phase transitions; the exact generation mechanism is dependent on the magnetic state of the material, and hence the generation and/or detection efficiency will show a change at a magnetic phase change.

It is well known that the velocity of ultrasound propagation in a material is related to its elastic properties [3]. For a single crystal the velocity of sound is dependent on the type of sound wave and the direction of propagation. For example, in a hexagonal crystal such as Gd, for propagation of sound along the c-axis two elastic constants can be measured from the velocities $v$ of longitudinal and shear waves; $C_{33} = \rho v_{\text{long}}^2$ and $C_{44} = \rho v_{\text{shear}}^2$, where $\rho$ is the density of the material [3]. The elastic constants will change as the properties of the sample change [3].

Elastic constant measurements require accurate determination of the velocity [4, 5]. Typical measurements are set up as shown in the inset of Figure 1; a sample has an ultrasonic transducer fixed to it and reflections from the opposite face are detected, with a typical echo pulse train shown (the peaks in the figure show the envelope of the MHz-frequency oscillation of the transducer). The time between echoes, $\Delta t$, needs to be measured, with the velocity given by

$$\Delta t = \frac{2L}{v} + \frac{\phi}{2\pi} \cdot \frac{1}{f}$$  \hspace{1cm} (1)

where $L$ is the sample thickness, $f$ is the frequency of the ultrasonic signals (typically in the MHz range) and $\phi$ is the phase change on reflection. This phase correction is typically small [4].
It is difficult to define a suitable measurement point for each echo. In the 1960s pulse echo overlap (PEO) was developed to measure the time between echoes [5]. In this method, two pulses are generated and the time between them altered such that the echoes overlap, giving a phase velocity accuracy of 2 parts in $10^4$, but requiring significant input from an operator. Recently, a digital equivalent using cross correlation has been developed [4], giving an accuracy of 1 part in $10^7$.

2. Automating the velocity measurement

The availability of faster processors has meant that automation of data analysis, through recording all the ultrasonic echoes and digital signal analysis on a PC, is now possible. We have built real-time data acquisition and analysis routines using LabVIEW [12].

Ultrasonic echoes measured using EMATs are often noise dominated, and this adds to the problems of identifying the start time of an echo for measurement of $\Delta t$. Simply measuring the maximum amplitude point in each echo could lead to the wrong part of the echo being chosen due to the presence of noise, and hence automation is difficult. Cross correlation, however, enables a clearer measurement of the time between echoes [4]. We have implemented a routine using the cross-correlation VI in LabVIEW; the first measured echo is windowed and scanned across the series of echoes by varying an offset time. The maxima in the cross correlated signal therefore correspond to the offset times whereby the first and later echoes are in best agreement, and this gives the transit time within the material. At present, this method does not take into account phase changes on each reflection of the signals; these are assumed to be negligible and will be investigated later. However, initial results show this to be a suitable technique for velocity measurements. The LabVIEW routine for data analysis also measures echo amplitudes for the first few echoes, and an amplitude for the noise level between echoes. These are used to measure the signal attenuation, and to measure the signal to noise for the first echo, which gives a good measure of the EMAT efficiency.

3. Measurements of Gd$_{64}$Sc$_{36}$ using contact and non-contact ultrasonic techniques

Gd-rich alloys show a competition between ferromagnetic order (from Gd) and other magnetic phases from the alloy materials, such as Sc, which shows helical magnetic ordering [14]. Gd$_{64}$Sc$_{36}$ has been shown to be a simple helimagnet from the Néel point down to the lowest temperatures, with the turn angle locked in place by 30 K [13]. The single crystal sample used for these experiments was grown at the Centre for Materials Science, University of Birmingham, UK, and used for the work presented in [14]. It has dimensions of approximately 4 mm. This material was chosen as the proof of concept sample test, as measurements had previously been made using standard manual PEO techniques and quartz transducers, with results available for comparison.

Measurements were taken using a Matec 6600 pulse generator system, capable of generating narrowband signals between 1 and 350 MHz given suitable transducers. Temperature control and magnetic field were provided by an Oxford Instruments superconducting magnet and cryostat system, in combination with a heater fitted to the experimental probe. Data collection was through a Tektronix oscilloscope with 350 MHz bandwidth communicating with a PC running LabVIEW. Contact measurements used an X-cut quartz transducer, generating 15 MHz longitudinal waves with waves propagating along the c-axis and the magnetic field also applied along this axis. This was used in pulse-echo mode to both generate and detect ultrasonic signals. Non-contact measurements used several different designs of EMATs, either to just detect signals (with the quartz transducer used for generation) or to both generate and detect ultrasound. The coil designs investigated included a simple spiral (pancake) coil, and several designs of differential coil [15]. The latter design uses a pair of coils and was intended to reduce noise by providing a reference signal away from the sample. The reference coil was kept as close to the original site as possible to experience the same magnetic field.
Ultrasonic signals were recorded over a range of temperatures and analysed using cross-correlation methods [4], giving a measure of $C_{33}$. Figure 2(a) shows $C_{33}$ as a function of magnetic field normalised to 1 for no applied field, measured using the differential coil EMAT to both generate and detect ultrasound, at a temperature of 90 K. Two main features can be seen; $C_{33}$ shows a very clear dip at around 1.6 T, corresponding to the change from a fan to a ferromagnetic phase [14]. A smaller feature, showing as a change in gradient, is also present at around 0.6 T; this corresponds to the change from distorted helix to fan phase, and is clearer when the applied field is along the $a$-axis. Despite the noise problems inherent in an EMAT measurement, application of suitable frequency filtering and the use of cross-correlation techniques have allowed analysis of the data.

![Figure 2. (a) Measurement of $C_{33}$ using a differential coil for generation and detection, at a temperature of 90 K. (b) Attenuation (black) and EMAT efficiency (red) measurements.](image)

The attenuation of the signal can also be measured, and is shown as the black trace in Figure 2(b). There is clearly a marked change in the signal attenuation around both phase transitions. With EMAT measurements a further measure of changes in the sample is possible through measuring the efficiency of generation and detection. As the exact generation and detection methods rely on the magnetic state of the material, it is to be expected that a change in this state will lead to a significant difference in the signal amplitudes being generated. For this measurement the efficiency is shown in red (right axis). In the region of phase changes the generation efficiency peaks, and it is clear that the fan phase is optimal for generation of signals. This will be investigated in more depth in a later publication.

![Figure 3. (a) $C_{33}$ measurement using a quartz transducer to generate and either quartz (black), a single EMAT coil (red) or a differential coil (green) to detect ultrasound. (b) shows a phase diagram created using each set of data (after [14]).](image)
Finally, we compare the results from several measurements using quartz transducers and/or EMATs with either a single coil for EMAT detection or a differential coil setup. Figure 3(a) shows normalised $C_{33}$ values from each of these detection transducers for generation using quartz, measured for zero applied magnetic field and over a temperature range from room temperature down to 10 K. As can be seen, the results all show a similar trend, with a slight discrepancy in temperature possibly due to an observed degradation of the temperature sensor calibration.

Figure 3(b) shows the summary of results from each experiment on a phase diagram, with the quartz measurements used as a reference and shown as dashed lines. Points correspond to $C_{33}$ measurements performed using different EMAT coils (for generation and detection) over a range of temperatures and magnetic fields. Red squares also show initial analysis of the EMAT efficiency. Again, very good agreement is shown between all methods.

4. Conclusions and future work

We have presented results from initial measurements using contact transducers, and using two different styles of EMAT coils for detection and generation of ultrasound. Results were compared for measurements of Gd$_{64}$Sc$_{36}$, and showed good agreement between the measurements. Results were analysed using a LabVIEW-based cross-correlation technique, which is a good step towards improving the problems caused by electrical noise in EMAT measurements.

Further improvement of the noise through the use of improved differential coils is anticipated. Also, other frequency-based techniques may be more appropriate for this echo measurement. Previous cross-correlation techniques have looked at the time difference between a reflection from a buffer rod and the first echo from a sample, and hence have fewer issues with phase changes on reflections. The fast Fourier transform of a string of echoes (as shown in Figure 1) gives a convolution of the frequency of the echo itself and the repeat pattern of the echoes [16], and is showing promise for analysing these measurements. We hope to extend these methods to further materials.

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