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SOUND PROPAGATION NEAR THE TRICRITICAL POINT OF $FeCl_2$ *

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The magnetically induced phase transitions, near the tricritical point of $FeCl_2$ were studied by acoustic velocity measurements. Longitudinal waves propagating along the (100) trigonal axis exhibit a critical shift in the velocity along the second order λ-line and anomalous change at the first order phase transitions. The phase diagram in the plane of temperature and applied magnetic field is constructed near the tricritical point.

1. INTRODUCTION

THE COUPLING of sound waves to critical fluctuations near phase transitions, gives rise to a pronounced attenuation and velocity shift [1]. These phenomena have been studied extensively mainly at 1st and 2nd order phase transitions. Here we report on the observation of a critical softening of the elastic stiffness near the tricitical point of a metamagnetic $FeCl_2$ [2–9]. $FeCl_2$ is an antiferromagnet with a layered type spin structure. There is a ferromagnetic exchange J_1 between the in-plane Fe^{2+}ions, and an antiferromagnetic exchange J_2 between adjacent planes, that is about twenty times weaker than J_1. The magnetic moments are aligned by a relatively high anisotropy perpendicular to the Fe^{2+} layers and along the trigonal axis of the crystal. It has been found that below the tricritical temperature $T_t \simeq 21$ K under an increasing applied magnetic field H_{app}, the crystal undergoes a first order transition to a paramagnetic state. The transition is at constant internal field and over a range of H_{app} and during the transition the crystal is a mixture of para- and antiferromagnetic domains. Above T_t the transition from the antiferromagnetic phase to the paramagnetic phase is of second-order type.

The magnetic phase diagram of $FeCl_2$ has been investigated by various groups using different techniques, e.g. magnetic measurements [3, 5], neutron diffraction [7] and magnetooptics [6, 8, 9]. All the experiments show a tricritical behaviour near the temperature $T \simeq 21$ K, similar to that observed for He^3-He^4 mixture [10]. The second-order λ-line in the $(M-T)$ phase diagram and the upper branch of the first order boundary lines, M^+, approach the tricritical point with different slope [7, 9]. This is inconsistent with the Landau-type tricritical behaviour [11]. On the other hand both first order boundary lines M^+ and M^- approach the tricritical point linearly [9], as expected by the Landau's theory and the logarithmic correction was found to be insignificant. These salient features of the phase diagram were observed also in our ultrasonic study.

2. EXPERIMENTAL RESULTS AND DISCUSSION

The crystal used in our measurements was grown by the Bridgman technique [4]. A sample of ~ 6 × 6 × 4 mm with its larger end faces parallel to the trigonal c-axis was cut out and carefully polished. The velocity measurements were carried out at 30 MHz using a phase comparison technique [13] with a resolution of 3 ppm. The temperature was controlled to better than 0.1 K.

Previous measurement [14] of the change in sound velocity as a function of temperature at $H_{app} = 0$ are shown in Fig. 1 for longitudinal waves propagating in the 001 and in the 100 direction. The critical effect at the Neel temperature T_N is much larger in the latter configuration. Such an anisotropy in the critical effect is expected for a layer type spin structure where the strain modulation of the exchange is anisotropic [12]

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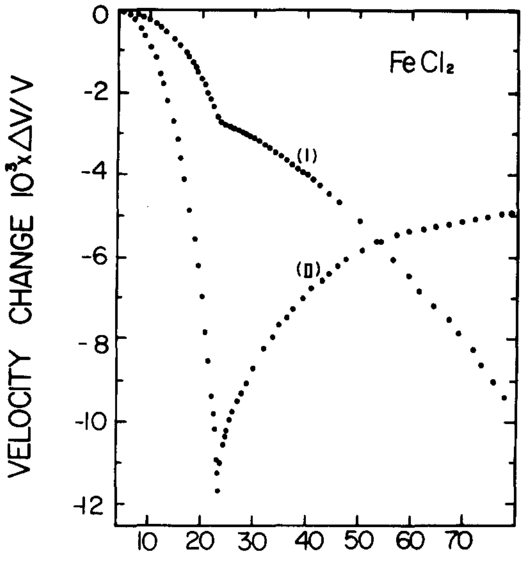

Fig. 1. Relative velocity change of a 30 MHz longitudinal sound wave measured as a function of temperature. (I) wave propagation along the (001) direction. See [14]. (II) wave propagation along the (100) direction.

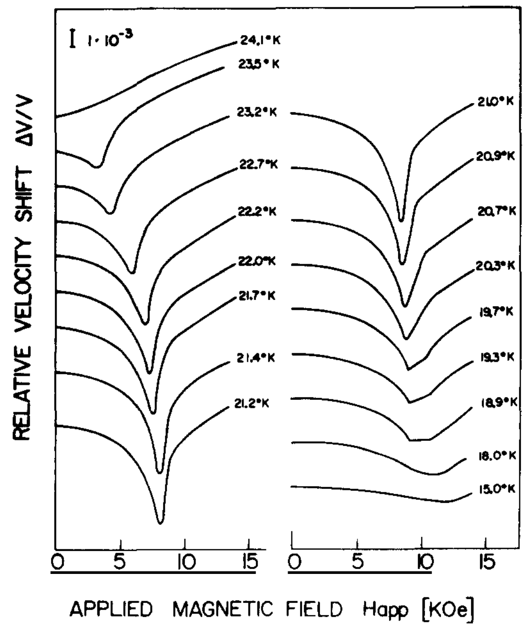

Fig. 2. Isotherms of a field dependent velocity. The relative velocity change DV/V of a 30 MHz longitudinal sound wave propagating in the (100) direction is measured as a function of a magnetic field applied along the trigonal (001) direction.

$\partial J_1/\partial \epsilon > \partial J_2/\partial \epsilon$, where ϵ is the strain. The present work on critical effect was therefore performed with longitudinal waves propagating in the 100 direction. Isotherm of the sound velocity as a function of the magnetic field H_{app} are shown in Fig. 2. In the isotherms at

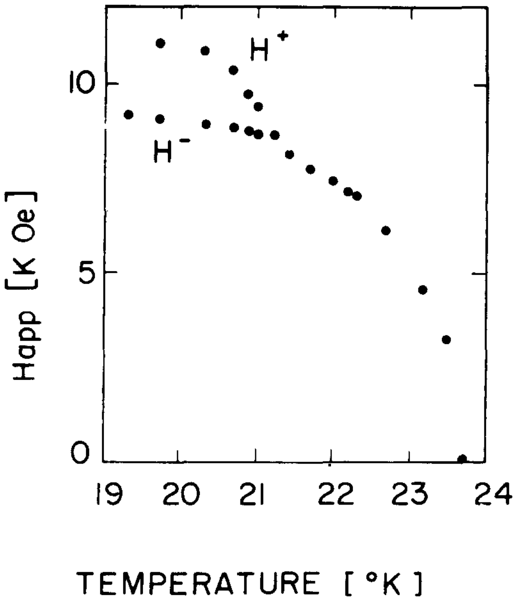

Fig. 3. Magnetic phase diagram of $FeCl_2$ based on the results given in Fig. 2.

temperature $T_N > T \geq T_t$ the observed minimum in the second velocity was assumed to occur at the critical field. This minimum becomes more pronounced as the temperature approaches $T_t = 21.2 \pm 0.1$ at which temperature the critical effect is maximum [15]. Below T_t the minimum becomes less pronounced and it is shallow and smeared below $T = 18$ K. The curves present two changes in slope at the minimum of the sound velocity and at a higher field. These two points were interpreted as the beginning and the end of the first order phase transition under the applied magnetic field. It should be mentioned that, although a coupling to the domain walls may effect the velocity in the mixed phase, different runs yielded the same results and no noticeable hysteresis was found upon increasing and decreasing the magnetic field.

The critical fields obtained from the various isotherms are given in Fig. 3. The first order boundary lines which are denoted here H^+ and H^- correspond to the first order boundary lines M^+ and M^- of the M–T phase diagram respectively. The relation between the applied field and magnetization M is given by:

$$H_{int} = H_{app} - 4\pi NM, \quad (1)$$

where H_{int} is the internal magnetic field and N is the demagnetization factor. It should be noted that the acoustic beam in these experiments was 1–2 mm wide and therefore the measurement involves only the central portion of the sample. Therefore N can be taken as constant in the measurement region and since the internal field remains constant through the first order transition [16] we have

$M^+ - M^- = (H^+_{app} - H^-_{app})/4\pi N.$

The first order boundary line should therefore have similar temperature dependence in the $H_{app} - T$ and in the $M-T$ diagram. As mentioned above M^+ and M^- were found to approach the tricritical point linearly. The results of Fig. 3 exhibit similar features.

The observed increase of the critical effect on sound velocity near the tricritical point may be examined in the spirit of the Landau theory of phase transitions. [11]. According to this theory near the phase transitions the free energy can be expressed in the form $G = G_0 + a\sigma^2 + b\sigma^4 + c\sigma^6$, where σ is the order parameter. The coefficients a, b, c depend on the temperature and magnetic field. At an ordinary critical point $a = 0$, $b > 0$ while at the tricritical point $a = b = 0$ and the free energy changes as the sixth power of the order parameter. It appears therefore that near the tricritical point larger fluctuations [17] are expected. The results remain valid for the local fluctuation of the order parameter and of related quantity like energy fluctuation although the modern theory shows that the correlation length diverges less rapidly on approaching a tricritical point than in approaching an ordinary critical point.

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REFERENCES

1. See e.g. GARLAND C.W., *Physical Acoustics* (edited by MASON W.P. & THRUSTON R.N.), p. 51. Academic Press, New York (1970);LUTHI B., MORAN T.J. & POLLINA R.J., *J. Phys. Chem. Solids* **31**, 1741 (1970); REHWALD W., *Adv. Phys.* **22**, 721 (1973).
2. GRIFFITHS R.B., *Phys. Rev. Lett.* **24**, 715 (1970).
3. JACOBS I.S. & LAWRENCE P.E., *Phys. Rev.* **164**, 866 (1967).
4. BIRGENEAU R.J., YELON W.B., COHEN E. & MAKOVSKY J., *Phys. Rev.* **B5**, 2607 (1972).
5. VETTIER C., ALBERTS H.L. & BLOCK D., *Phys. Rev. Lett.* **31**, 1414 (1973).
6. CHEN E.Y., DILLON J.F. & GUGGENHEIM H.J., *Magnetism and Magnetic Materials* – 1973 *AIP Conference Proceedings, No.* 18 (Edited by GRAHAM C.D. Jr. & RHYNE J.J.). American Institute of Physics, New York (1974).
7. BIRGENEAU R.J., SHIRANE G., BLUME M. & KOEHLER W.C., *Phys. Rev. Lett.* **33**, 1098 (1974).
8. GRIFFIN J.A., SCHNATTERLY S.E., FARGE Y., REGIS M. & FONTANA M., *Phys. Rev.* **B10**, 1960 (1974).
9. GRIFFIN J.A. & SCHNATTERLY S.E., *Phys. Rev. Lett.* **33**, 1576 (1974).
10. GRAF E.H., LEE D.M. & REPPY J.D., *Phys. Rev. Lett.* **19**, 417)1967).
11. LANDAU L. & LIFSHITZ E.M., *Statistical Physics*, p. 452. Pergamon Press, London (1958); *Collected Papers of L. Landau* (Edited by TER HAAR D.), p. 193. Gordon & Breach, New York (1967).
12. LUTHI B., MORAN T.J. & POLLINA R.J., Reference [1].
13. MORAN T.J. & LUTHI B., *Phys. Rev.* **187**, 710 (1969).
14. GORODETSKY G., SHAULOV A., VOLTERRA V. & MAKOVSKY J., *Phys. Rev.* **B13**, 1205 (1976).
15. Slightly different results for T_c were reported in [3–9]. Our result is found to be in agreement with that of [7].
16. WYATT A.F.G., *J. Phys.* **C1**, 684 (1968).
17. ACHIAM Y. & IMRY Y., *J. Phys.* **C10**, 39 (1977).