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Ultrasonic Attenuation near the Spin-Flip Temperature in Chromium

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Measurements of the attenuation coefficient of longitudinal ultrasonic waves have been made on a single crystal of chromium near the spin-flip order-order transition. Measurements were made for sound propagation along a ⟨100⟩ axis with the crystal in the normal state and after magnetic field cooling through the Néel point in two different geometries. Considerable differences in behavior were found between the different magnetic geometries and also upon applying a large magnetic field near the spin-flip temperature. A mechanism involving switching of the polarization of the spin density waves at domain boundaries is proposed to account for the results. The relaxation time associated with this switching is estimated to be 10^{-10} sec.

INTRODUCTION

It has been recognized in the last few years that ultrasonic attenuation studies near phase transitions can give valuable information about phase-change mechanisms as well as enable the determination of the critical indices which characterize the transitions.[1–5] Of particular interest in this connection is the study of chromium, since it is now well established that below 312 °K chromium is an antiferromagnet of the spin-density-wave type. Between about 122 and 312 °K the spin density waves are transversely polarized. As chromium is cooled through the spin-flip temperature T_F at 122 °K, it undergoes a first-order phase transition, and the spin polarization changes from transverse to longitudinal. The variation of the sound attenuation near the Néel temperature T_N was previously studied by O'Brien and Franklin[6] and by Luthi, Moran, and Pollina,[7] who observed a large peak in the absorption at T_N. We have performed similar measurements near the order-order transition at T_F and report the results in this paper.

EXPERIMENTAL PROCEDURE

The measurements were made on two polycrystalline samples of lengths 0.941 and 1.010 cm, and one single crystal of chromium of length 0.815 cm. The polycrystalline samples were spark cut from a bar supplied by MRC and annealed in a vacuum of better than 10^{-6} mm Hg for about 10 h at 1250 °C. A grain structure etch showed that the samples consisted of several grains from 1 to 5 mm in diameter. The resistance ratios of the samples after annealing were both 130. The single-crystal sample, prepared from a bar supplied by Aremco, was spark cut so that sound propagation was along a ⟨100⟩ axis and then given the same annealing treatment as the polycrystals. The resistance ratio of the single crystal was 50.

X-cut quartz transducers of 10-MHz fundamental frequency were used to generate longitudinal acoustic waves. They were bonded to the samples with indium in the case of the polycrystals and with Araldite Epoxy for the single crystal. Changes in the attenuation were measured by the pulse-echo technique. The first echo was gated, passed through a PAR Box Car integrator and logarithmic amplifier, and recorded on the Y axis of an X-Y recorder. Temperature was measured with a copper-constantan thermocouple varnished directly to the specimen. The signal from the thermocouple was recorded on the X axis of the X-Y recorder.

The sample was contained in the inner can of a two-can cryostat. By evacuating the space between the cans and putting exchange gas in the inner can, it was possible to vary the temperature of the sample slowly and uniformly. It was also possible to "field cool" the sample, i.e., cool the sample through the Néel point at 312 °K while in a strong magnetic field (46 kG) provided by a superconducting solenoid. If the magnetic field is along a ⟨100⟩ axis of the crystal, this will ensure the establishment of a spin density wave with a single wave vector $\vec{Q}$ parallel to the same ⟨100⟩ axis throughout the crystal.[8] Experiments were performed in which there was no field cooling, in which the sample was field cooled so as to establish the $\vec{Q}$ vector parallel to the sound propagation direction $\vec{q}$, and with $\vec{Q}$ normal to the propagation direction.

Measurements were made by cooling or warming the sample through the spin-flip transition at rates between 5 and 10 min/°K. No significant change in the attenuation behavior was observed when this rate was decreased to 30 min/°K. When measurements were made over an extended temperature range, some variation in the attenuation was observed that was ascribed to variations in the transducer bond. The bond variation with temperature was sufficiently slow that the effect could be ignored when considering a limited temperature range near T_F.

Experiments were also carried out to investigate the effect of a magnetic field applied normally to $\vec{Q}$ on a previously field-cooled crystal.

EXPERIMENTAL RESULTS

The results obtained with the polycrystalline samples in the non-field-cooled and parallel field-cooled cases showed the same qualitative features as the more detailed results obtained with the single crystal, and so discussion will be confined to the latter case. Figure 1 shows typical X-Y recorder tracings obtained for temperatures near the

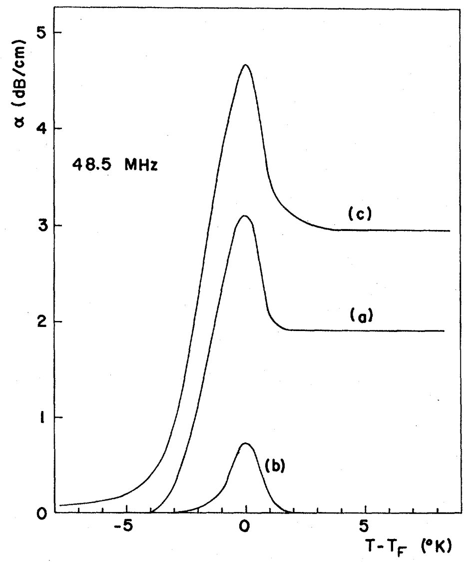

FIG. 1. Attenuation as a function of temperature near T_F: (a) with no field cooling at T_N; (b) cooling field parallel to the sound propagation direction; (c) cooling field normal to the sound propagation direction.

spin-flip transition in the three cases: curve (a) with no field cooling at the Néel point; curve (b) with $\vec{Q}$ parallel to $\vec{q}$; and curve (c) with $\vec{Q}$ normal to $\vec{q}$. In all three curves illustrated the frequency was close to 50 MHz. A curve similar to curve 1(a) has recently been published.[9] Two distinct features are apparent: a sharply peaked rise in attenuation near the transition and a step change in the background attenuation above and below T_F. The size of the step is strongly dependent on the magnetic state of the chromium, being absent when $\vec{Q}$ is parallel to $\vec{q}$, large when $\vec{Q}$ is perpendicular to $\vec{q}$, and intermediate in size when the crystal is not in a single-$\vec{Q}$ field-cooled state. In contrast, the peak at T_F is not so strongly dependent on the magnetic state. These observations indicate that two separate mechanisms are responsible for the two effects, a conclusion supported by the observation that the size of the step is strongly amplitude dependent: When the amplitude of the 50-MHz sound wave was reduced by 20 dB, the step was found to decrease from 2.9 to 1.2 dB/cm. But the attenuation peak did not show any such amplitude dependence.

The occurrence of such a marked amplitude dependence in the size of the step masked the essential frequency dependence, but values obtained for frequencies from 30 to 90 MHz appeared consistent with $\Delta\alpha = \alpha(T > T_F) - \alpha(T < T_F)$ being proportional to the frequency squared.

By making continuous recordings of attentuation from temperatures below T_F to above T_N and then going back to below T_F in a different field-cooled state, it was found that the background attentuation at $T < T_F$ was the same for all three possible field-cooling conditions. The differences in the attenuation step can thus be ascribed to differences in the attenuation at temperatures between T_F and T_N.

In order to study the sharp rise in attenuation near T_F, we made detailed measurements for the situation where $\vec{Q}$ is parallel to $\vec{q}$. This avoids the difficulty of separating out the background jump observed for other magnetic states. Measurements were made at frequencies between 50 and 110 MHz, and at each frequency results similar to curve (b) in Fig. 1 were found. The magnitude of the peak attenuation was found to be proportional to the square of the frequency, as shown in Fig. 2. No theory is available to indicate the behavior that might be expected in the ordered state, particularly near a first-order transition such as this. Nevertheless, we have attempted to analyze the behavior in terms of critical indices, as is customarily done in discussing the behavior of the paramagnetic state near a Néel or Curie point. By drawing a log-log plot of attenuation versus $T - T_F$ at temperatures near T_F, it was found possible to fit an equation of the form $\alpha(T) = A|T - T_c|^{-\lambda}$ in the region 0.7–3 °K above the transition. The value of the exponent thus found was approximately the same at all frequencies with the value $\lambda = 2.5$ (± 0.2). A similar range of temperatures could be fitted with the same exponent for $T < T_F$.

In order to further reduce the possible spin-wave states of the crystal, additional experiments to investigate the effect of an applied magnetic field while the sample was in the transversely field-cooled state ($\vec{Q}$ normal to $\vec{q}$) were also performed. The field was applied along a ⟨100⟩ axis normal to $\vec{Q}$, thereby forcing the spins to lie in a [010] direction normal

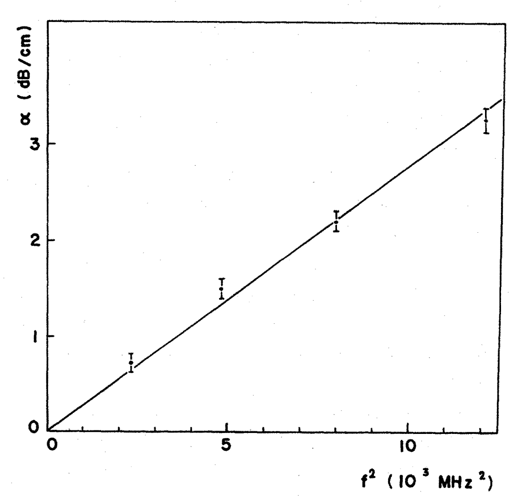

FIG. 2. Maximum attenuation at T_F as a function of frequency squared with the sample field cooled parallel to the propagation direction.

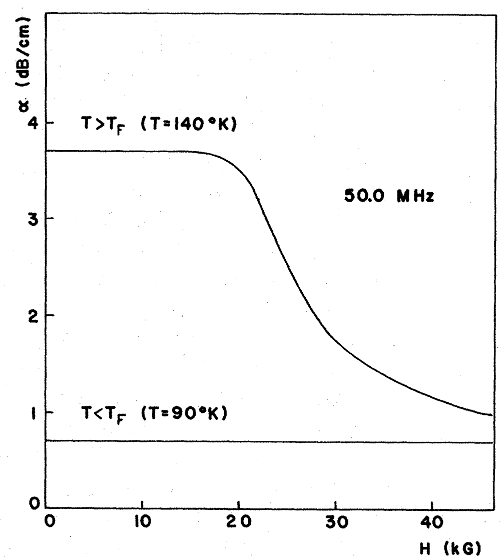

FIG. 3. Attenuation as a function of magnetic field applied parallel to the propagation direction, sample field cooled normal to the propagation direction.

to both $\vec{Q}$ and $\vec{H}$ ($T_F < T < T_N$) or parallel to $\vec{Q}$ but normal to $\vec{H}$ ($T < T_F$).[10] When $T > T_F$, there are two possible types of transversely polarized domains, one with the spins normal to $\vec{Q}$ and $\vec{q}$, one normal to $\vec{Q}$ but parallel to $\vec{q}$. The application of the magnetic field will cause one of the domain types to grow at the expense of the other depending on whether $\vec{H}$ is parallel or perpendicular to $\vec{q}$. Experiments were performed with both orientations of field with very similar results. Figure 3 shows curves obtained when $\vec{H}$ is parallel to $\vec{q}$. As the field is increased the attenuation is reduced, saturating in high fields at the value found in zero field when $T < T_F$. Application of H while $T < T_F$ does not change the existing single-domain structure with spin parallel to $\vec{Q}$. Figure 3 shows the attenuation in this situation to be independent of the field. No change in the field-dependent behavior was noted either when the amplitude of the signal was reduced by 5dB, or when the temperature was varied between 130 and 170 °K.

The effect of the magnetic field on the attentuation peak at T_F was also investigated by setting the field at a high value (46 kG) and sweeping the temperature through the transition. The results are shown in Fig. 4 for a frequency of 50 MHz. Curve (a) is the result when $H = 0$ with a single $\vec{Q}$ normal to $\vec{q}$. In (b) $H = 46$ kG, $\vec{H}$ is normal to both $\vec{Q}$ and $\vec{q}$; and in (c) $H = 46$ kG, $\vec{H}$ is normal to $\vec{Q}$ but parallel to $\vec{q}$.

DISCUSSION

Attenuation Step

Two possible mechanisms have been considered to account for the attenuation step at T_F: scattering of the sound at magnetic domain boundaries or interaction between the sound wave and the spin density wave leading to a rotation of the spin polarization within a domain. The domain structure arises in the non-field-cooled case since there are three equivalent possible directions for $\vec{Q}$. Between T_N and T_F each $\vec{Q}$ has two possible spin polarizations (transverse) giving a total of six possible types of domain. Below T_F there is only one (longitudinal) polarization possible for each $\vec{Q}$ giving three possible types of domain. However, when the sample is field cooled to produce a single $\vec{Q}$, there are two possible types of domain above T_F and only one below. When $\vec{Q}$ is parallel to $\vec{q}$, the two domains ($T > T_F$) are identical with respect to the sound wave, and so it is not to be expected that any switching of the polarization would occur either at the boundary or within a domain. This is in accord with the observed behavior: $\alpha(T > T_F) = \alpha(T < T_F)$. When the single $\vec{Q}$ is normal to $\vec{q}$, the two domains are no longer symmetrical, and one can expect that the alternate dilation and compression of the crystal as the sound wave passes will cause some switching of the polarization, and hence some attenuation. If this attenuation is due to an interaction within the bulk of either, or both, types of domain, one would expect the transversely field-cooled state to exhibit $\frac{3}{2}$ as much attenuation as the non-field-cooled state, assuming equal occurrence of the three possible $\vec{Q}$ directions in the latter case. This is close to the observed factors of between 1.27 and 1.56. It is not so clear what the expected ratio would be if the interaction is taking place primarily at the domain boundaries. However, if it is assumed that boundaries between different $\vec{Q}$ domains are ineffective or small in number, and that the dominant scatterers are boundaries between different polarizations with the same $\vec{Q}$, then the same factor of $\frac{3}{2}$ is arrived at.

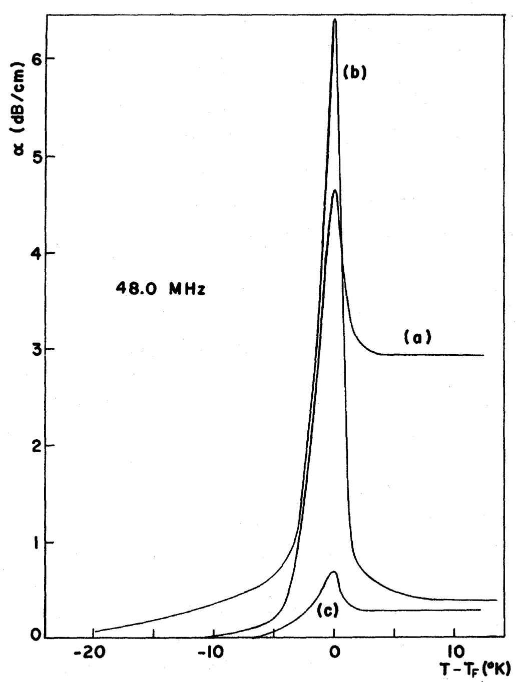

FIG. 4. Attenuation as a function of temperature near T_F, sample field cooled normal to the propagation direction: (a) with no applied field; (b) $H = 46$ kG, $\vec{H}$ normal to $\vec{Q}$ and $\vec{q}$; (c) $H = 46$ kG, $\vec{H}$ normal to $\vec{Q}$ but parallel to $\vec{q}$.

A test between these two models is provided by the magnetic field dependence of the attenuation. If the interaction is a bulk effect, at least one of the two field orientations perpendicular to $\vec{Q}$ that were used would drive the entire crystal into a single interacting domain with the result that the attenuation would either remain constant or increase with increasing field. But the observed effect for both field orientations is to suppress the "step attenuation" entirely. While not conclusive evidence, this is consistent with a domain wall attenuation mechanism since the domain walls will tend to disappear as the increasing field forces the crystal into a single-$\vec{Q}$ single-polarization state. It therefore appears that the cause of the step in attenuation is scattering of the sound at boundaries between different spin polarization domains within a single-$\vec{Q}$ state. This interpretation is supported by the observation that the peak found when T is swept through T_F is the same when H is zero and when $H = 17$ kG ($\vec{H}$ parallel to $\vec{q}$). If the spins were switched at a lower field, the peak at T_F would then be expected to be much smaller, as is discussed below. The magnetic field dependence shows little hysteresis, thus supporting the argument of Werner *et al.*[10] for the existence of pinning sites which locally fix the direction of the spin. As the field is increased, the size of energetically unfavorable domains decreases and may disappear. As the field is decreased, the pinning sites become free to exert their influence and the domains grow again, returning to the same equilibrium distribution in zero field.

Further evidence is provided by Steinitz's[11] observation of the field dependence of the attenuation in the same geometry. With a crystal of higher resistance ratio than reported on here, he found that the step attenuation was suppressed by a smaller magnetic field. This indicates that in the purer crystal there are fewer pinning sites, leaving the domains freer to change size in response to an applied magnetic field.

The amplitude dependence of the step attenuation is then interpreted in this model as being due to an activation energy associated with the spin pinning sites. A large amplitude wave will be more likely to free the domain boundary from the pinning site. This then gives an amplitude-dependent attenuation in a manner similar to the well-known case of attenuation by dislocations pinned at points along their length.

Attenuation Peak

A clue as to the mechanism involved in the attenuation peak near the spin-flip transition is also provided by the magnetic field experiments whose results are illustrated in Fig. 4. In curve (b) the field is applied so as to establish a single domain with $\vec{Q}$ normal to $\vec{q}$, spin parallel to $\vec{q}(T > T_F)$. The peak attenuation is then very much greater than in curve (c) when the single domain has both $\vec{Q}$ and spin normal to $\vec{q}$. In both cases the state when $T < T_F$ is with spin parallel to $\vec{Q}$ and normal to $\vec{q}$. The effective sound-absorbing mechanism is thus associated with the transition in which the spin changes from parallel to normal to $\vec{q}$, and not so strongly when the spin moves from one normal position to the other. Now Steinitz *et al.*[12] have shown that there is a discontinuity in the crystal lattice dimension parallel to $\vec{q}$ in the former case, and none (or very small) in the latter case. Near the transition, the alternate compression and dilation of the lattice caused by the sound wave then provides a mechanism which can drive regions of the crystal through the transition leading to a relaxation attenuation. The small residual peak in curve (c) could be ascribed to a small number of spins remaining pinned in the unfavorable direction, as suggested by Steinitz *et al.* to account for the small observed discontinuity in the corresponding lattice dimension. This mechanism would also account for the observed peak in the longitudinal field-cooled crystal [curve 1(b)], for in that case there is again a discontinuity in the lattice dimension parallel to $\vec{Q}$[12] and $\vec{q}$ at T_F, thus permitting the transition to be driven by a compressional wave. The magnitude of the peak observed for longitudinal field cooling would not necessarily be the same as for transverse field cooling. By comparing the results when the entire crystal is in an interacting domain configuration [curve 1(b) and curve 4(b)], it can be seen that the longitudinal peak is much less than the transverse peak.

For both the peak and step attenuation, a mechanism has been proposed involving a disturbance of the domain configuration and a relaxation toward equilibrium. For such a relaxation process, the attenuation coefficient is generally given by the expression

$$\alpha = \alpha_0 \, \omega^2 \tau / (1 + \omega^2 \tau^2), \tag{1}$$

where ω is the angular frequency and τ is the relaxation time. If $\omega\tau \ll 1$, this leads to a frequency-squared dependence of the attenuation. We observed such a dependence at frequencies up to 90 MHz. If we take this to mean that $\omega^2 \tau^2 < 0.1$, this puts an upper bound on the relaxation time, $\tau < 10^{-9}$ sec. On the basis of measurements of internal friction, Munday *et al.*[13] have previously estimated an upper limit of 10^{-5} sec for the relaxation time associated with absorption by transversely polarized domains.

One can also express the attenuation coefficient due to a relaxation process in terms of ΔY, the maximum change in Young's modulus Y as the wave passes. The resulting attenuation is[14]

$$\alpha = \frac{\Delta Y}{Y} \frac{1}{2v} \frac{\omega^2 \tau}{1 + \omega^2 \tau^2} , \tag{2}$$

where v is the sound velocity. Street *et al.*[14] have determined values of $\Delta Y/Y$ near T_F for a field-cooled polycrystalline specimen with the measuring direction parallel to the field cooling. From their results we may estimate a value of $\Delta Y/Y = 10^{-2}$ for the peak at T_F. This leads to a value of the relaxation time $\tau \sim 10^{-10}$ sec for the process associated with the longitudinal field-cooled peak.

In the absence of similar Young's modulus data for a transverse field-cooled specimen, it is not possible to make a similar estimate of the relaxation time for the transverse field-cooled peak or for the step attenuation mechanism.

From the magnetic field required to suppress the domain structure (Fig. 3), it is possible to estimate the energy associated with the domains. The change in energy when a volume V of the crystal switches its polarization from parallel to an applied field H to normal to H is $\Delta\chi H^2 V$, where $\Delta\chi$ is the difference in susceptibility of the two polarizations. Published measurements of susceptibility do not distinguish between these two types of domain orientation, but we may estimate a value of $\Delta\chi = 10^{-7}$ emu/g.[15] From Fig. 3, a field of about 25 kG is needed to suppress the domain. Hence the energy associated with the boundaries between domains of different transverse polarization but same vector $\vec{Q}$ is estimated as -6×10^{-6} J/g. This is expected to be sample dependent, varying with the number of pinning points, contributing 1–2% to the latent heat at the flip transition which is estimated by Street *et al.*[16] and Steinitz *et al.*[12] to be 4×10^{-4} or 5×10^{-4} J/g.

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Two-Spin-Wave Resonances and the Analyticity of the *t* Matrix in the Heisenberg Ferromagnet*

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We consider the scattering of two spin waves in a simple cubic Heisenberg ferromagnet with nearest-neighbor interaction at zero temperature. The analytic properties of the integrals relevant to the bound-state problem are examined and the solutions to the bound-state conditions are located in the complex energy (ω) plane for the first time. It is found that, as a function of ω, there are two Riemann sheets near the bottom of the two-spin-wave band. For total wave vector $\vec{q}$ less than the threshold value for bound states, the existence of the d-wave resonant states discovered by Boyd and Callaway is reaffirmed. Furthermore, we confirm the observation of Boyd and Callaway that no s-wave scattering resonance exists.

I. INTRODUCTION

It has been shown by Dyson[1] that two spin waves in a Heisenberg ferromagnet interact via an attractive potential which increases with the total wave vector $\vec{q}$ of the pair. Bound or resonant states of two spin waves may exist for large $\vec{q}$. The two-spin-wave bound-state conditions have been obtained by Hanus[2] and by Wortis.[3] Later, Boyd and Callaway[4] and Silberglitt and Harris[5] obtained the same conditions by different methods. Wortis[3] has discussed the bound states in great detail. He found that, for total wave vector $\vec{q}$ larger than a threshold, there are three bound states below the two-spin-wave band, among which two states are degenerate if $\vec{q}$ is in the [111] direction. Boyd and Callaway[4] derived an expression for the two-spin-wave scattering cross section for $\vec{q}$ along the [111] direction, which they resolved into two partial-wave components, s and d waves. They showed that both s- and d-wave bound states exist, and that the d-wave state is doubly degenerate, while the s-wave state is nondegenerate. Furthermore, they pointed out that the d-wave bound states connect to resonant scattering states in the band as q passes the threshold, while the s-wave states do not show resonant behavior.[6] Looking into the singularities of the two-spin-wave t matrix, Silberglitt and Harris[5] obtained a two-spin-wave spectrum which agrees with the results of Hanus,[2] Wortis,[3] and Boyd and Callaway.[4] They also gave a physical reason why there is a d-wave resonance but no resonant s state, and investigated the effect of the d-wave resonance on the single-spin-wave spectrum.

The purpose of this paper is to reaffirm the existence of the d-wave resonance and to confirm the nonexistence of the s-wave resonance by examining the analytic properties of the integrals relevant to the bound-state problem and locating the solutions to the s- and d-wave bound-state conditions in the complex energy plane. We find