Landau model for the elastic properties of the quasi-one-dimensional antiferromagnetic compound CsNiCl₃

G. Quirion and T. Taylor
Department of Physics and Physical Oceanography, Memorial University, St. John’s, Newfoundland, Canada A1B 3X7

M. Poirier
Regroupement Québécois sur les Matériaux de Pointe, Département de Physique, Université de Sherbrooke, Sherbrooke, Québec, Canada J1K 2R1

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We present a Landau model that accounts for the unusual elastic properties of the quasi-one-dimensional antiferromagnetic compound CsNiCl₃. The model’s predictions are tested using published strain measurements and recent high resolution sound velocity measurements realized as a function of temperature and pressure. In particular, we derive an analytical expression which indicates how the low temperature dependence of the elastic constants is related to the critical order parameters. A close inspection of the temperature dependence of $C_{33}$ indicates that the value of the critical exponent $\beta$ associated with the order parameter of the elliptical state below $T=4.10$ K is $\beta=0.33$. The obtained value agrees with the expected critical behavior of CsNiCl₃ in the absence of an external magnetic field (conventional three-dimensional XY criticality). Using sound velocity measurements under hydrostatic pressures up to 7.0 kbar, we also derive the pressure-temperature phase diagram of CsNiCl₃. With increasing pressure, we observe that the critical temperatures $T_{N_1}$ and $T_{N_2}$ increase at a rate of $dT_{N_1}/dP=0.17$ K/kbar and $dT_{N_2}/dP=0.065$ K/kbar, respectively. Finally, taking advantage of the magnetoelastic coupling in the paramagnetic state, we could estimate the pressure dependence of the interchain exchange coupling along the $c$ axis, $dJ_{c}/dP=0.45$ kbar. All these results seem to indicate that the Ising-type anisotropy and the quasi-one-dimensional character of CsNiCl₃ are both enhanced with pressure.

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I. INTRODUCTION

Over the past decades, there has been a considerable interest in the properties of quasi-one-dimensional antiferromagnetic materials with a generic chemical formula $ABX_3$, where $B$ represents magnetic ions such as Ni, Cu, Co, Cr, and Fe.¹ In this class of hexagonal insulators, the magnetic ions form linear chains along the $c$ axis which are weakly coupled antiferromagnetically in the $a$-$b$ plane. Thus, these geometrically frustrated spin systems, due to the antiferromagnetically coupling on a triangular lattice, are expected to exhibit new types of magnetic ordering. Among this class of quasi-one-dimensional antiferromagnet materials, CsNiCl₃ is certainly the compound that stimulated the most interest. This particular spin-1 quasi-one-dimensional Heisenberg antiferromagnet is one of the first compounds to show experimental evidences of the Haldane gap.² Moreover, it has still been used as a prototype system for the investigation of quantum fluctuations³–⁵ on the properties of quasi-one-dimensional antiferromagnets. The Hamiltonian describing the magnetic properties of these systems is

$$H = -2J_0 \sum_{i} \mathbf{S}_i \cdot \mathbf{S}_{i+1} - J_\parallel \sum_{i \neq j} \mathbf{S}_i \cdot \mathbf{S}_j - D \sum_{i} (S^z_i)^2 - \mathbf{H} \cdot \sum_{i} \mathbf{S}_i,$$

where $J_0$ and $J_\parallel$ are the intrachain and interchain nearest-neighbor antiferromagnetic exchange interactions, respectively, and $D$ the single-ion anisotropy. In the case of CsNiCl₃, considering that $J_0=-16.6$ K, $J_\parallel=-0.26$ K,³,⁶ and $D=0.048$ K,⁷,⁸ the system has an easy-axis magnetization along $\hat{c}$ (Ising-type anisotropy, $D>0$) while CsMnBr₃ (Ref. 9) is known to favor an in-plane magnetization (XY-like anisotropy, $D<0$). Due to the competition between these interactions, these quasi-one-dimensional antiferromagnet materials show interesting magnetic ordering at low temperatures. In particular, there has been a considerable amount of work toward the determination of the magnetic phase diagram of CsNiCl₃ in order to demonstrate that it does indeed possess a new type of multicritical point.¹⁰–¹⁴ The nature of this multicritical point, where one first-order boundary connects with three second-order lines, is now well established due to the theoretical work realized by Plumer et al.¹⁰ For a magnetic field $\mathbf{H}$ applied along the hexagonal $c$ axis, they have shown that the free energy can be expressed as

$$F = C_s S^2 - C_\parallel S^2 \cos^2 \theta + C_\perp S^2 \sin^2 \beta + 2B_2 S^4 \sin^4 \beta$$

$$+ \frac{1}{2} A_0 m^2 + \frac{1}{2} B_1 S^4 + \frac{1}{2} B_2 m^4 - mH.$$

Here, the free energy depends on three order parameters, $S$, $\beta$, and $\theta$, while $m$ represents the uniform magnetization induced by an external magnetic field $\mathbf{H}$ applied along the $c$ axis. Thus, these order parameters can be used to describe the different ordered phases observed at low temperatures. Minimization of the free energy [Eq. (2)] with respect to the order parameters reveals that there are four magnetic phases characterized as

$P$, paramagnetic state, $S=0$;
As shown by Plumer et al. (Ref. 15), the theoretical predictions agree very well with the experimental observations.

\[ L, \text{ linear state, } S_\parallel = S (\theta = 0), \]
\[ S_\perp = 0 (\beta = 0); \]

\[ E, \text{ elliptical state, } S_\parallel = S (\theta = 0), \]
\[ S_\perp = S \sin \beta (\beta \neq 0); \]

\[ SF, \text{ 120° structure state, } S_\parallel = 0 (\theta = \pi/2), \]
\[ S_\perp = S \sin \beta (\beta = \pi/4); \]

where \( S_\parallel = S \cos \theta \) and \( S_\perp = S \sin \beta \). These different ordering states are all clearly identified in Fig. 1 where we present the magnetic phase diagram of CsNiCl\textsubscript{3}. Even if the model includes a large number of adjustable parameters most of them can be easily adjusted using the measured critical values \( T_{N_1}, T_{N_2}, T_m, H_m, \) and \( H_{SF} \). As shown by Plumer et al.,\textsuperscript{15} this model reproduces remarkably well the experimental magnetic phase diagram of CsNiCl\textsubscript{3}. Thus, Eq. (2) certainly represents a good starting point for the elaboration of an extended model which can be used to analyze the properties of CsNiCl\textsubscript{3}, in particular its elastic properties. Even if elastic measurements have been extensively used in order to obtain the magnetic phase diagram of CsNiCl\textsubscript{3},\textsuperscript{11,16,17} no serious attempt has been made to analyze the unusual temperature dependencies observed at low temperatures. Consequently, in order to validate the proposed model, we realized complementary ultrasonic velocity measurements as a function of temperature and pressure. Moreover, these new measurements allow us to estimate the pressure dependence of the in-plane and out-of-plane exchange coupling parameters as well as to determine the pressure-temperature phase diagram of CsNiCl\textsubscript{3}.

**II. EXPERIMENT**

The CsNiCl\textsubscript{3} single crystal used in this investigation was grown by the Bridgman method and had approximately the shape of a parallelepiped with a length of 8.9 mm along the crystalline \( c \) axis and roughly 2.5 mm along the transversal directions. All measurements presented in this work have been obtained using 30 MHz piezoelectric transducers in order to generate and detect longitudinal acoustic waves. The relative variations in the sound velocity (\( \Delta V/V \)) were measured using a high-resolution pulsed acoustic interferometer. For measurements realized under pressure, the transducer-sample assemblage was inserted in a Cu-Be pressure cell filled with a 3-methyl-1-butanol fluid acting as the pressure-transmitting medium. As well, mounted close to the sample, a small wire of lead was used as a pressure manometer during the experiment. Consequently, the loss of pressure related to the thermal contraction of the different components of the pressure cell was carefully monitored and taken into account in the data analysis.

**III. ELASTIC PROPERTIES OF CsNiCl\textsubscript{3} AT AMBIENT PRESSURE**

Ultrasonic techniques\textsuperscript{11,17,18} have been used extensively as a mean to obtain the magnetic phase diagram of CsNiCl\textsubscript{3}, surprisingly, the values of its elastic constants are still undetermined. For that reason, we present in Table I the velocity of a few acoustic modes. These measurements, realized at room temperature, have been obtained by measuring the time of flight between echoes. Knowing that CsNiCl\textsubscript{3} has a hexagonal crystal structure (\( D_{6h}^2 \) space group)\textsuperscript{1} and an estimated density of \( \rho = 3700 \) kg/m\textsuperscript{3}, it is straightforward to obtain the elastic constants reported in Table I. What is certainly more interesting is that there are, so far, no models that adequately account for the unusual temperature dependence of its elastic properties at low temperatures. Thus, in order to facilitate our analysis, we extended the temperature range of investigation down to 2 K using longitudinal waves propagating along and perpendicular to the hexagonal axis (\( c \) axis). In Fig. 2(a), we first present the temperature dependence of \( C_{33} \) obtained from the velocity measurements along the \( c \) axis. According to these results, the onset of the three-dimensional linear antiferromagnetic ordering corresponds to a small slope variation at \( T_{N_1} = 4.75 \) K while the elliptical magnetic ordering is easily identified by a pronounced anomaly at \( T_{N_2} = 4.31 \) K. The strong anisotropy of the different magnetic orders is confirmed by results obtained for \( C_{11} \) which we present in Fig. 2(b). In this case, we observe no sudden variation in either \( C_{11} \) or in its temperature dependence at \( T_{N_1} \), while the second transition at \( T_{N_2} \) is easily identified by a rapid drop. For the first time, we show that both elastic constants, \( C_{11} \) and \( C_{33} \), still change rapidly in the low temperature elliptical magnetic state. A question that one might ask is whether or not this temperature dependence, just below \( T_{N_2} \), is somehow related to that of an order parameter. As the alterations of the properties of a crystal are normally accom-

**TABLE I. Sound velocities and elastic constant measurements obtained at room temperature for CsNiCl\textsubscript{3}.

<table>
<thead>
<tr>
<th>Mode</th>
<th>Velocity (m/s)</th>
<th>Elastic Constant (N/m\textsuperscript{2})</th>
</tr>
</thead>
<tbody>
<tr>
<td>( V_{[100]} )</td>
<td>6140</td>
<td>( C_{11} = 13.9 )</td>
</tr>
<tr>
<td>( V_{[001]} )</td>
<td>4080</td>
<td>( C_{33} = 6.16 )</td>
</tr>
<tr>
<td>( V_{T,1} )</td>
<td>2500</td>
<td>( C_{44} = 2.31 )</td>
</tr>
<tr>
<td>( V_{T,2} )</td>
<td>3100</td>
<td>( C_{66} = 3.56 )</td>
</tr>
<tr>
<td>( C_{12} )</td>
<td>6.78</td>
<td></td>
</tr>
</tbody>
</table>

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point group symmetry, the exact elastic term $F_{el}$ expression can easily be derived.\(^{19}\) However, considering that there is no change in the point group symmetry, the shear strain components $(e_1, e_2, e_3)$ can be omitted. Moreover, in order to reduce the number of adjustable parameters, we also neglect any terms associated with the elastic constants $C_{13}$ and $C_{12}$. Therefore, the elastic energy $F_{el}$ considered here reduces to

$$F_{el}(e_i) = \frac{1}{2}C_{11}(e_1^2 + e_2^2) + \frac{1}{2}C_{33}e_3^2 + P(e_1 + e_2 + e_3),$$

where $C_{ik}$ represent the bare elastic constants of the high temperature phase. We also take into account the contribution associated with the application of an external pressure $P$. In general, in the presence of any external stresses, contributions such as $-\sum_{i} \sigma_i e_i$, where $\sigma_i$ represents the applied stress tensor, must be considered. For a hydrostatic pressure $P$, $\sigma_i = -P$ whenever $i=1,2,3$, and zero for $i=4,5,6$ (shear strains). As one can see, taking into account the pressure in the free energy does not increase the number of adjustable parameters. However, this allows us to calculate the pressure dependence of the critical temperatures. Finally, the lower order coupling interaction terms that one must necessarily consider are

$$F_e(S, S_\perp, e_i) = \left[ g_{s_1}(e_1 + e_2) + g_{s_3}e_3 \right] S^2$$

$$+ \left[ g_{\beta_1}(e_1 + e_2) + g_{\beta_3}e_3 \right] S_\perp^2.$$  

Naturally, it is straightforward to show that all terms considered in Eq. (3) are indeed invariant with respect to the transformations of the space group $P6_3/mmc$ as well as with respect to time reversal. Thus, the Landau free energy represented by Eqs. (4)–(6) should give a proper phenomenological framework that can be used to derive the temperature dependence of many thermodynamic quantities. In particular, this approach should provide a useful link between the order parameters and the experimental data. From the minimization of the free energy, Eq. (3), with respect to $e_i$, we obtain the expressions for the spontaneous strains $e_i(Q)$. At ambient pressure these relations correspond to

$$e_1(S, S_\perp) = e_2(S, \beta) = -\frac{g_{s_1}S^2 + g_{\beta_1}S_\perp^2}{C_{11}},$$

$$e_3(S, S_\perp) = -\frac{g_{s_3}S^2 + g_{\beta_3}S_\perp^2}{C_{33}}.$$  

It is possible, from the same free energy, to derive the elastic constants by using\(^{20}\)

$$C_{mn} = \frac{\partial^2 F}{\partial e_m \partial e_n} = \frac{\partial^2 F}{\partial Q \partial e_m} \left( \frac{\partial^2 F}{\partial Q^2} \right)^{-1} \frac{\partial^2 F}{\partial e_n \partial Q},$$

where $Q$ represents the order parameter associated with a specific magnetic ordering. Consequently, we obtain

$$C_{11} = C_{11} - \frac{g_{s_1}^2}{B}, \quad T_{N_2} < T < T_{N_1},$$

$$C_{11} = C_{11} - \frac{g_{\beta_1}^2}{4B_2}, \quad T < T_{N_2},$$

FIG. 2. Relative variation of the elastic constants as a function of temperature, (a) and (b) show $C_{33}$ and $C_{11}$, respectively. Symbols represent the data points while the continuous line corresponds to the theoretical prediction obtained using Eqs. (10) and (11). The dashed line is obtained by taking into account higher coupling terms, Eqs. (12) and (13). The inset in (a) shows $C_{33}$ as a function of the reduced temperature $\tau = 1 - T/T_{N_1}$ on a log-log scale. The value of the critical exponent obtained from the fit is $\beta = 0.33 \pm 0.02$.  

panied by a response in its elastic properties, ultrasonic velocity measurement is certainly a convenient way to detect any types of phase transitions, which explains why this type of measurement is frequently used. Moreover, since changes in the elastic properties are also subject to symmetry constraints imposed by the nature of the phase transition itself, these same measurements can be used to analyze the nature of the order parameter. Thus, using simple symmetry arguments, we derive here a phenomenological Landau model that can be used to account for the observed changes in the elastic properties of CsNiCl$_3$. In practice, if one takes into account the coupling between the order parameter $Q$ and the strains $e_i$, the free energy $F(Q, e_i)$ can be separated in three distinct constituents,

$$F(Q, e_i) = L(Q) + F_{el}(e_i) + F_e(Q, e_i),$$

where $L(Q)$ is the Landau-type free energy expressed in terms of a power series of the order parameter $Q$. $F_{el}(e_i)$ represents the elastic energy associated with the deformations, and $F_e(Q, e_i)$ takes into account the strains $e_i$ which are coupled to the order parameter $Q$. In the case of CsNiCl$_3$, the Landau-type free energy\(^{15}\) has already been derived and used to account for the magnetic phase diagram of CsNiCl$_3$. Thus, the Landau free energy $L(Q)$ represented by Eq. (2) certainly represents a valid starting point and does not require any further justifications. In the absence of an external magnetic field, considering that $\theta = 0$ and defining $S_\perp = S \sin \beta$, Eq. (2) can be rewritten as

$$L(S, S_\perp) = A_S S^2 + C_{11} S^2 + 2B_S (S_\perp^2 - S^2) S_\perp^2.$$  

We see that the Landau free energy depends on two order parameters, $S$ which is associated with the linear phase while $S_\perp$ drives the system into an elliptical magnetically ordered state at $T_{N_2}$. Knowing that CsNiCl$_3$ belongs to the hexagonal
on the elastic properties of CsNiCl$_3$ in the basal plane. Thus, by imposing $C_d$cedence of both model certainly fails to account for the temperature dependence associated with critical phenomena. In fact, if a non-mean-field temperature dependence is used for the order parameters, it is possible to obtain a better agreement. Unfortunately, the resolution of the data presented in Fig. 3 does not allow for a detailed analysis of the critical behaviors.

Even if the proposed model reproduces the changes at the phase transitions, it fails to account for the variation of $C_{33}$ and $C_{11}$ below $T_N$. For that reason, we now consider what could be the contribution of higher coupling terms. Considering that there are no changes in $C_{11}$ at $T_N$, the only higher coupling terms that we now consider are the biquadratic terms such as $\gamma_1(e_1+e_2)^2S_1^2 + \gamma_2 e_1^2 S_{1.1}^2$. Naturally, these additional terms change all analytical relations derived so far, however, in this particular case, these corrections are small and can be ignored. The most significant influence associated with those coupling terms is to account for the temperature dependence of the elastic constants at low temperatures. When the higher coupling terms are taken into account, a direct numerical comparison indicates that the elastic constants are well approximated by

$$C_{11}^* = C_{11}, \quad T_{N_2} < T < T_{N_1},$$

$$C_{11}^* = C_{11} - \frac{8g_{S_1}}{B}, \quad T < T_{N_2}$$

for $C_{11}$ and $C_{33}$. Considering that our experimental results indicate that there are no changes on $C_{11}$ at $T_{N_1}$, we must impose that the coupling parameter $g_{S_1} = 0$. This outcome is also well supported by thermal expansion measurements realized by Rayne et al.\textsuperscript{16} According these thermal expansion data, reproduced in Fig. 3 for further comparison, measurements perpendicular to the $c$ axis show no anomaly at $T_{N_2}$ [see Fig. 3(b)]. This clearly indicates that the ordering of the magnetic moments along the $c$ axis below $T_{N_1}$ has no effect on the elastic properties of CsNiCl$_3$ in the basal plane. Thus, by imposing $g_{S} = 0$, the proposed model is consistent with the fact that both phase transitions are easily identified on $C_{33}$ while $C_{11}$ decreases only at $T_{N_2}$. However, this simple model certainly fails to account for the temperature dependence of both $C_{11}$ and $C_{33}$ below $T_{N_2}$. As we will see shortly, this discrepancy can be removed by considering higher coupling terms. However, at this point it is still convenient to compare the experimental observations and the analytical predictions derived so far. In particular, the measured variations of $C_{11}$ and $C_{33}$ at the phase transitions provide an accurate way to determine the only adjustable parameters ($g_{B_1}/C_{11}=8.04 \times 10^{-7}$, $g_{S_3}/C_{33}=4.43 \times 10^{-7}$, $g_{B_2}/C_{33}=-1.85 \times 10^{-6}$) where the elastic constants correspond to those listed in Table I. The other parameters are given in Ref. 7 and have been determined prior to this work using data obtained for the magnetic phase diagram alone. Based on these numerical values, the continuous lines in Fig. 2 correspond to the model’s predictions whenever fourth order coupling terms are considered. The pertinence of the proposed model can then be tested by comparing the calculated thermal expansions [using relations (7) and (8)] with the data published by Rayne et al.,\textsuperscript{16} see Fig. 3. There is no doubt that the proposed model has some merit since it captures the general trend with the proper magnitudes. Naturally, considering that the continuous lines in Fig. 3 represent mean-field predictions, we cannot expect to reproduce the temperature dependence associated with critical phenomena. In fact, if a non-mean-field temperature dependence is used for the order parameters, it is possible to obtain a better agreement. Unfortunately, the resolution of the data presented in Fig. 3 does not allow for a detailed analysis of the critical behaviors.

Even if the proposed model reproduces the changes at the phase transitions, it fails to account for the variation of $C_{33}$ and $C_{11}$ below $T_N$. For that reason, we now consider what could be the contribution of higher coupling terms. Considering that there are no changes in $C_{11}$ at $T_N$, the only higher coupling terms that we now consider are the biquadratic terms such as $\gamma_1(e_1+e_2)^2S_1^2 + \gamma_2 e_1^2 S_{1.1}^2$. Naturally, these additional terms change all analytical relations derived so far, however, in this particular case, these corrections are small and can be ignored. The most significant influence associated with those coupling terms is to account for the temperature dependence of the elastic constants at low temperatures. When the higher coupling terms are taken into account, a direct numerical comparison indicates that the elastic constants are well approximated by

$$C_{11}^* = C_{11}, \quad T_{N_2} < T < T_{N_1},$$

$$C_{11}^* = C_{11} - \frac{8g_{S_1}}{B} + \gamma_3 S_{1.1}^2, \quad T < T_{N_2}$$

$$C_{33}^* = C_{33} - \frac{g_{S_3}}{B}, \quad T_{N_2} < T < T_{N_1},$$

$$C_{33}^* = C_{33} - \frac{g_{S_3}}{B} + \gamma_3 S_{1.1}^2, \quad T < T_{N_2}$$

According to these relations, the temperature dependence of $C_{11}$ and $C_{33}$ in the elliptical magnetically ordered state is directly related to the square of the order parameter $S_{1.1}$. Naturally, we cannot expect to reproduce the observed temperature dependence if we limit our analysis to a mean-field approach. As shown by the thermal expansion data (Fig. 3), the temperature dependence associated with critical phenomena is certainly relevant in the case of CsNiCl$_3$. Thus, if a non-mean-field temperature dependence is adopted, the elastic constants below $T_N$ can be well fitted using $S_{1.1} = S_{1.1}^0(1 - T/T_N)^\beta$ as long as the critical exponent $\beta = 0.33 \pm 0.02$. Here, the critical temperature $T_{N_2}$ is accurately determined by taking the temperature corresponding to the minimum on the elastic constant curve. The power law behavior below $T_{N_2}$ is better appreciated in the inset of Fig. 2(a) which shows the variation of $C_{33}$ as a function of the reduced temperature $\tau = 1 - T/T_{N_2}$ on a log-log plot. The data indicate that the power law dependence is well defined over at least one decade close to $T_{N_2}$. Moreover, the value of $\beta$ obtained in this work agrees very well with those obtained from neutron
scattering,21 NMR,22 and specific heat measurements.12 All these results are consistent with the theoretical expectation that, at low magnetic fields, this critical line belongs to the XY universality class with a value of $\beta=0.35$. Finally, we point out that the deviations between the model’s prediction and the experimental results above both phase transitions are probably related to low-dimensional fluctuations. Similar fluctuation effects are observed on the elastic properties of the quasi-one-dimensional system such as CuGeO$_3$.23,24 In the case of CsNiCl$_3$, considering that only one-third of the magnetic moments are effectively ordered in the linear state, we still expect large fluctuations until the full three-dimensional character is established below $T_{N_2}$.

IV. SOUND VELOCITY MEASUREMENTS UNDER PRESSURE

In this section, we focus on the results obtained at high pressures. These results have been obtained using exclusively longitudinal waves propagating along the hexagonal axis. This particular direction has been chosen because it allows an easy determination of both critical temperatures ($T_{N_1}$ and $T_{N_2}$). The most significant results obtained at low temperatures are presented in Fig. 4(a) where the curves, corresponding to different pressures, have been arbitrarily shifted relative to each other for an easier comparison.

As shown in Fig. 4(a), both anomalies move to higher temperatures as pressure increases. Using this series of measurements, we directly derive the pressure-temperature phase diagram of CsNiCl$_3$ that we present in Fig. 4(b). Our investigation as a function of pressure indicates that the critical temperatures increase at a rate of $dT_{N_1}/dP=0.17$ K/kbar and $dT_{N_2}/dP=0.065$ K/kbar, respectively. These variations agree well with recent heat capacity measurements realized under pressure.25 In that case, the observed variations correspond to $dT_{N_1}/dP=0.19$ K/kbar and $dT_{N_2}/dP=0.087$ K/kbar, respectively. These values can also be compared to the predictions derived from our model (for our calculation, $a=400$ was used). By including terms associated with pressure in Eq. (5), we can estimate the pressure dependence of the critical temperatures. According to the proposed model

\[
\frac{dT_{N_1}}{dP} = \frac{gS_1}{aC_{33}} = 0.11 \text{ K/kbar,}
\]

\[
\frac{dT_{N_2}}{dP} = \frac{dT_{N_1}}{dP} + \frac{B}{2aB_2} \left( \frac{2g\rho_1}{C_{11}} + \frac{8g\rho_2}{C_{33}} \right) = 0.06 \text{ K/kbar,}
\]

which compare remarkably well with the experimental results. We obtain as well that the pressure dependence of the critical temperatures is essentially determined by the relative strength of the coupling terms and the elastic constants. In the case of CsNiCl$_3$, since $g\rho_1$ is the only coupling term which is negative, the coupling between $e_3$ and $S_1$ reduces the rate at which $T_{N_2}$ changes and contributes to favor the linear magnetic ordering relative to the elliptical state as pressure increases.

The Hamiltonian describing the magnetic properties of CsNiCl$_3$ is given by Eq. (1) and as mentioned previously $J_1$ and $J_\perp$ represent the intrachain and interchain nearest-neighbor antiferromagnetic exchange interactions, respectively, and $D$ is associated with the single-ion anisotropy. The competition between $D$ and $J_\perp$ gives rise to two magnetic phases separated by only a few degrees ($T_{N_1}$ and $T_{N_2}$) at $H=0$ T. The single-ion anisotropy $D$ has a tendency to favor the linear antiferromagnetic phase $L$ which is observed below $T_{N_1}$. Because of the magnetic frustration produced by the antiferromagnetic interaction $J_\perp$ on the triangular lattice, the spins conserve some degrees of freedom in the basal plane. At lower temperature, this frustration is eventually overcome and the remaining degrees of freedom are then frozen in the elliptical phase $E$ which takes place below $T_{N_2}$. Thus, the fact that the temperature range of the linear phase $L$ increases with pressure might be an indication that the single-ion anisotropy $D$ increases more rapidly than $J_\perp$ with respect to pressure. This is also supported by experimental results obtained on a series of CsNi$_{1-x}$M$_x$Cl$_3$ alloy compounds, ($M =$Co, Fe, and Mg).26 On the one hand, when magnetic Ni ions are substituted by nonmagnetic Mg ions, a dilution of single-ion anisotropy is achieved and a decrease in $T_{N_1}$ is observed. Naturally, this substitution also influences $J_\perp$ which explains why $T_{N_2}$ is also reduced, by roughly the same

FIG. 4. Comparison of the relative variation of the sound velocity (a) measured as a function temperature at constant pressures, using longitudinal waves propagating along the hexagonal axis of CsNiCl$_3$, (b) shows the pressure-temperature phase diagram derived from sound velocity measurements.
amount as $T_{N_1}$. On the other hand, if Ni ions are substituted by Co ions, which have a stronger Ising-type anisotropy (increasing the global anisotropy $D$), a rapid increase in $T_{N_1}$ is observed with only a small reduction in $T_{N_2}$. Finally, let us mention that recent specific heat measurements on CsNiCl$_3$ also indicate that the Ising-type anisotropy ($D > 0$) increases with pressure.

So far, we have limited our discussion to how the transverse nearest-neighbor coupling $J_{\perp}$ and the single-ion anisotropy $D$ evolve under pressure without any references to the parallel coupling $J_\parallel$. Fortunately, in the case of quasi-one-dimensional magnetic systems such as CsNiCl$_3$, it is possible to determine the magnitude of this coupling by measuring the temperature dependence of the sound velocity in the paramagnetic state. As shown in Fig. 5, pressure modifies the temperature dependence of $C_{33}$ even well above the critical temperatures. It has been shown that the unusual temperature dependence on $C_{33}$, corresponding to a plateau centered around 40 K for CsNiCl$_3$, can be associated with a strong magnetoelastic coupling along the $c$ axis. This unusual phenomenon, which has been the object of previous ultrasonic investigations, can be described using a classical model where the magnetoelastic coupling is introduced via the modulation of the parallel exchange integral $J_\parallel$ as a function of the spin separation along the $c$ axis. When this coupling is considered, a softening in $C_{33}$ is expected to take place at a temperature corresponding to $2J_\parallel$. Thus, in order to clearly identify the contribution due to the magnetoelastic coupling alone, we need to subtract the anharmonic elastic contribution responsible for the temperature dependence of all elastic constants. Here, we approximated that contribution by a linear extrapolation of the high temperature dependence ($T > 150$ K). One can see the results of this manipulation in the inset of Fig. 5, which illustrates the influence of pressure on the magnetoelastic coupling alone. We see that the magnitude of the magnetoelastic coupling decreases with increasing pressure. This indicates that, as the crystal stiffens under pressure, the influence of the magnetoelastic becomes less preeminent. What is more interesting is that the temperature $T_s$, corresponding to the temperature for which we observe the maximum softening, increases with pressure. Considering that this temperature scales with $2J_\parallel/k_B$, we can deduce that the parallel exchange integral $J_\parallel/k_B$ increases at a rate of $dJ_\parallel/dP = 0.45$ K/kbar. Assuming that $T_{N_2}$, rather scales with $J_\perp$, our measurements indicate that the one-dimensional character of CsNiCl$_3$, $|J_\perp/J_\parallel| \ll 1$, is reinforced under pressure.

It is interesting to point out that, above the magnetic phase transitions, $C_{11}$ displays a temperature dependence which is similar to that of $C_{33}$. As shown in Fig. 6, $C_{11}$ increases rapidly as we approach the magnetic phase transition from above. In the instance of $C_{33}$ this is associated with magnetoelastic effects along the $c$ axis. Thus, it is reasonable to assume that the unusual temperature of $C_{11}$ is related to magnetoelastic effects taking place in the basal plane. After subtracting the anharmonic elastic contribution, we present in the inset of Fig. 6 the variation that could be associated with the magnetoelastic coupling in the basal plane. However, here the maximum softening is observed at 5.2 K, which is an order of magnitude larger than the interchain nearest-neighbor antiferromagnetic exchange interaction $J_\parallel = -0.26$ K. Thus, even if the temperature dependence of the observed softening is analog to that of a magnetoelastic coupling, we note an important discrepancy between the temperature of the maximum softening and $J_\perp$. Thus, compared to the magnetoelastic model derived by Fivez, it might be interesting to verify if this difference can be explained by considering two-dimensional fluctuations in the basal rather than one-dimensional fluctuations along the $c$ axis.

V. CONCLUSIONS

In this paper, we presented a Landau model that accounts for the pressure dependence of $T_{N_1}$ and $T_{N_2}$, as well as the temperature dependence of the strains and the elastic constants. In particular, we show how the temperature dependence of $C_{11}$ and $C_{33}$ can be used to obtain the value of the critical exponent $\beta$ associated with the elliptical state. There has been considerable interest over the past years about the properties of CsNiCl$_3$. In particular, as shown in Fig. 1, the magnetic phase diagram of CsNiCl$_3$ shows two phase boundaries at low field and a tetracritical point at $(T_m, H_m)$. Based...
on a scaling analysis, the critical behavior at low field should be that of a conventional three-dimensional XY system and our analysis of the temperature dependence of $C_3$ is consistent with that prediction. Moreover, Kawamura et al. also predict that the critical behavior close to the tetracritical point should rather correspond to that of a $\eta=3$ chiral fixed point ($\beta=0.28$) while the criticality along the phase boundary at higher field is of $\eta=2$ chiral universality characterized by exponents. Thus, our theoretical and experimental analyses of the elastic properties of CsNiCl$_3$ clearly demonstrate that sound velocity measurements provide a convenient way to study this type of critical phenomenon. Now that we clearly establish how the temperature dependence of $C_3$ is related to the order parameter, similar measurements should give us the opportunity to carry out a detailed investigation about how the critical exponent $\beta$ evolves as a function of a magnetic field.

Our experimental results also show that the critical temperatures $T_{N1}$ and $T_{N2}$ increase with pressure at a rate of $dT_{N1}/dP=0.17$ K/kbar and $dT_{N2}/dP=0.065$ K/kbar. Moreover, taking advantage of the strong magnetoelastic coupling observed along the $c$ axis, we derived the pressure dependence of the parallel exchange integral $(dJ_z/dP=0.45$ K/kbar). As the parallel coupling increases more rapidly than the other two couplings ($D$ and $J_z$), our results indicate that the one-dimensional character of CsNiCl$_3$ is reinforced under pressure. This indicates that CsNiCl$_3$ is probably not a suitable candidate for the investigation of a pressure-induced crossover regime between quasi-one-dimensional to two-dimensional regime. However, there might be other $ABX_3$ compounds suitable for this type of investigation.

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