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Abstract

We report measurements of the elastic moduli as a function of temperature (5-300) K and magnetic field (0-2 T) for the Zintl ferromagnet $Yb_{14}MnSb_{11}$, which is believed to be a rare example of an under-screened Kondo lattice. The elastic moduli measured below the Curie temperature in this complex ferromagnet exhibit unusual lattice stiffening that is independent of the magnetic field and can be adequately modeled using the Landau theory.

Introduction

The soft Zintl ferromagnet $Yb_{14}MnSb_{11}$ belongs to the family of "14-1-11" compounds, which are known to exhibit a wide range of unusual magnetic and electronic transport properties [1,2,3,4]. Isostructural to the Zintl compounds $Ca_{14}AlSb_{11}$ and $Ca_{14}MnB_{11}$, [5,6] $Yb_{14}MnSb_{11}$ crystallizes in a tetragonal lattice of the space group $I4_1/acd$ with 208 atoms per unit cell. The complex structure combined with the large

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number of atoms per unit cell and the heavy atomic masses of Yb and Sb leads to a relatively low thermal conductivity and the compound has recently attracted attention for its potential as a p-type thermoelectric material at high temperatures [7]. In addition, $Yb_{14}MnSb_{11}$ displays highly unusual magnetism, an understanding of which is expected to provide insights into the complex physics of ferromagnetic semiconductors, the key components of spintronic devices [8,9,10]. It has been suggested that the $Yb_{14}MnSb_{11}$ is a rare example of an under-screened Kondo lattice. [8, 11, 12] The Yb^{2+} ions in the $Yb_{14}MnSb_{11}$ are non-magnetic due to their filled 4f shells ($4f^{14}$), and X-ray magnetic circular dichroism measurements (XMCD) indicate a divalent Mn^{+2} (d^5) configuration. [13] The magnetic Mn atoms in the $Yb_{14}MnSb_{11}$ are found to be located at unique crystallographic sites within the $MnSb_4$ tetrahedra which results in a nearest neighbor Mn-Mn distance of approximately 10 Å. [1, 8] Hence, the ferromagnetic order in the $Yb_{14}MnSb_{11}$, whose onset temperature is T_c = 53 K, is attributed to RKKY interaction between the localized Mn 3d moments mediated via conduction holes from the Sb 5p bands. [14,15]

In this paper, we present and discuss the temperature dependence of the elastic moduli of $Yb_{14}MnSb_{11}$ below T_c. Changes in Young's modulus with magnetization have been recognized over one hundred years ago in materials such as iron and nickel. These materials show a sudden drop in the elastic modulus below T_c, which is referred to as the "delta-E effect" (ΔE effect). This strongly field-dependent change in the elastic moduli occurs because of domain wall motion under stress [16]. In contrast, the temperature- and field dependence of the elastic moduli obtained on $Yb_{14}MnSb_{11}$, a soft ferromagnet, show no significant ΔE effect below the Curie temperature. Instead, a lattice stiffening is

observed below T_c, similar to the temperature dependence of the elastic moduli of the ferromagnetic Mott insulator $YTiO_3$ [17]. The classical Landau theory [18,19,20] is utilized to model the thermodynamics of this second order magnetic phase transition.

Experimental technique

$Yb_{14}MnSb_{11}$ polycrystals and single crystals were synthesized at Oak Ridge National Laboratory, using a molten metal flux as first reported by Fisher *et al.* [2]. Resonant Ultrasound Spectroscopy (RUS) was used to measure the elastic moduli as a function of temperature and magnetic field. RUS is based on the measurement of the resonances of a freely vibrating body [21,22,23]. The mechanical resonances can be calculated for a sample with known dimensions, density, and elastic tensor. In a RUS experiment, the mechanical resonances of a freely vibrating solid of known shape are measured, and a nonlinear optimization procedure is used to model the measured lines with the calculated spectrum. The RUS data reported here were carried out as a function of temperature (5-300 K) and magnetic field (0-2 Tesla) on two polycrystalline rectangular parallelepipeds of approximately 2 x 2 x 5 mm^3, using a custom designed probe that can be inserted in a commercial Quantum Design Physical Properties Measurement System (PPMS). In addition, RUS data have been collected for a shard of single crystalline $Yb_{14}MnSb_{11}$. The shape and size of this single crystal did not allow us to cut an oriented parallelepiped (i.e. a parallelepiped with all faces perpendicular on the tetragonal crystal axes), which is essential to obtain quantitative values of the single crystalline moduli. However, RUS measurements can give important information even when it is not possible to obtain an absolute value for the elastic constants: anomalous thermodynamic behavior will be reflected in the temperature-dependence of the resonant frequencies, and these can be

measured regardless of sample shape or symmetry. The magnetization data used in the Landau analysis were obtained through standard susceptibility measurements in a Quantum Design Magnetic Properties Measurement System (MPMS).

Results and Discussion

Figure 1 shows the elastic moduli *vs*. temperature for polycrystalline $Yb_{14}MnSb_{11}$ measured for B = 0T and B = 2T. If untextured, polycrystalline materials are elastically isotropic, and have only two independent elastic moduli, the longitudinal modulus c_{11} (Figure 1(a)), and the shear modulus c_{44} (Figure 1(b)), which are related to respectively the longitudinal and transverse sound velocity in the material. Normal elastic behavior can be modeled with the so-called Varshni function [24], which shows a gradual increase of c_{11} with decreasing temperature, leveling off at low temperatures. The elastic response of $Yb_{14}MnSb_{11}$ clearly deviates from Varshni behavior: whereas both the longitudinal and shear modulus increase with decreasing temperature, a rather abrupt stiffening is observed below the Curie temperature (T_c = 53 K), which is not affected by the application of a magnetic field. A similar behavior is observed in single crystalline $Yb_{14}MnSb_{11}$, as shown by the temperature-dependence of a resonant frequency measured for a shard of single crystalline $Yb_{14}MnSb_{11}$ (Figure 2). Since the square of the resonant frequencies is directly proportional to the elastic moduli, the substantial increase in frequency observed below T_c reflects a stiffening of the elastic moduli, and confirms the observations for the polycrystal. The mean velocity of sound v_m can be calculated from the measured values of the polycrystalline moduli c_{11} and c_{44} using $v_L = \sqrt{\frac{c11}{\rho}}$ and $v_T = \sqrt{\frac{c44}{\rho}}$ and $v_m = (\frac{1}{3}[\frac{2}{v_T^3} + \frac{1}{v_L^3}])^{-1/3}$ [25]. The values of v_m calculated from the longitudinal and

transverse components of the elastic moduli varies from 1920 m/sec at 300K to 1942 m/sec at 10 K, resulting in a 1% increase in the mean velocity of sound in polycrystalline $Yb_{14}MnSb_{11}$ at temperatures below T_c. The Debye temperature (Θ_D) calculated from the mean velocity of sound is $\Theta_D \approx 186$ K at room temperature, using $\Theta_D = \frac{h}{k_B}\left[\frac{3qN\rho}{4\pi M}\right]^{1/3} v_m$ [25] in fair agreement with estimates from specific heat measurements which yielded $\Theta_D \approx (160 \pm 10)$ K. [1]

We track the temperature dependence of the elastic constants by calculating them as the second derivatives of the free energy with respect to strain at constant temperature. Although the RUS frequencies used in the experiment probe the adiabatic elastic constants, nominally obtained as second order derivatives of the *internal energy* in respect with the strain, from general thermodynamic considerations one can show that the ratio of the two sets of coefficients is proportional with the ratio of the heat capacities of the material measured at constant strain and constant force respectively (generalized Reech relation), which in a solid is close to unity.

In a magnetic system, however, the temperature-dependence of the elastic moduli is expected to be sensitive to changes in the magnetization of a material, because a change in the alignment of magnetic moments with changing temperature will affect the magnetic contribution to the free energy. We incorporate these aspects in a theoretical model based on the phenomenological Landau theory of phase transitions [18,19, 20] which permits an analysis of the temperature dependence of the elastic moduli in the presence of ferromagnetic ordering.

In this approach, the free energy F of a ferromagnet is written as a sum of terms that depend only on the order parameter, which is the spontaneous magnetization M, contributions from the elastic strain e, independent of M, and a contribution from the coupling between the order parameter M and the elastic strain e. In a crystal with tetragonal symmetry, the free energy thus constituted is written as:

$$F = -\frac{a}{2}M^2 + \frac{b}{4}M^4$$
$$+\frac{1}{2}c_{11}(e_{xx}^2 + e_{yy}^2) + c_{12}e_{xx}e_{yy} + c_{13}(e_{xx}e_{zz} + e_{yy}e_{zz}) + \frac{1}{2}c_{33}e_{zz}^2 + \frac{1}{2}c_{44}(e_{yz}^2 + e_{xz}^2) + \frac{1}{2}c_{66}e_{xy}^2$$
$$+M^2[d_1(e_{xx}^2 + e_{yy}^2) + d_2e_{xx}e_{yy} + d_3e_{zz}(e_{xx} + e_{yy}) + d_4e_{zz}^2 + d_5(e_{yz}^2 + e_{zx}^2) + d_6e_{xy}^2 + d_7e_{zz}$$
$$+d_8(e_{xx} + e_{yy})] \qquad (1)$$

The above expression for the free energy includes only even powers of M to reflect the invariance under time reversal. The easy direction of spontaneous magnetization is the c axis in this case, which belongs to the point group A_{2g} representation of 4/*mmm*. The latter terms result in the development of a strain under magnetization, preserving the tetragonal symmetry.

To simplify the calculation, we use a reduced generic version of Eq. 1, written as:

$$F = -\frac{a}{2}M^2 + \frac{b}{4}M^4 + H(e_i, e_j) + M^2G(e_i, e_j) \qquad (2)$$

The first two terms in equation (2) represent the pure magnetic contribution to the free energy. In the vicinity of the critical temperature, the phenomenological parameter a assumes a temperature dependence of the form $a(T) = a_0(T_c - T)$, with a_0 a temperature independent constant. The second phenomenological parameter b is also temperature independent. $H(e_i, e_j) = \frac{1}{2}\sum_{ij}^{6} c_{ij}e_ie_j$ describes the elastic part with the strain in the

absence of M. The components of the strain tensor are indexed by i, j =1 to 6 such that 1 ≡ xx; 2 ≡ yy; 3 ≡ zz; 4 ≡ yz; 5 ≡ zx; 6 ≡ xy [26]. Finally, $G(e_i, e_j)$ results from the coupling between elastic strain and the magnetization, with d_i the magnetoelastic coupling constants. Following the Slonczewski-Thomas model, [27] the magnetization is assumed to vary quasi-statically as a function of the deformation, such that it maintains its equilibrium value. This is obtained by differentiating the free energy in respect to the magnetization. Therefore,

$$\frac{\partial F}{\partial M} = 0 \tag{3}$$

which leads to:

$$-aM + bM^3 + 2GM = 0 \tag{4}$$

The non-trivial solution for magnetization is:

$$M_0^2 = \frac{a - 2G}{b} \tag{5}$$

We note that on account of the explicit temperature dependence assumed for a, as discussed above, the equilibrium magnetization acquires, in a first order approximation where the contribution from the elastic deformation is neglected, the same behavior.

When Eq. (5) is substituted back in F, the free energy becomes a function of e_i, e_j. The dependence is explicit and also implicit through M. This new form of the free energy is labeled $\hat{F}$ following the Slonczewski-Thomas notation. The elastic coefficients are calculated as the second derivative with respect to a certain displacement of $\hat{F}$. Using our simplified expression, Eq. (2), the elastic constants are:

$$c_{11} = \partial^2 \hat{F} / \partial e_1^2 = \frac{\partial}{\partial e_1}\left(\frac{\partial \hat{F}}{\partial e_1}\right) = \frac{\partial}{\partial e_1}[(-aM_0 + bM_0^3 + 2M_0 G)\frac{\partial M_0}{\partial e_1} + \frac{\partial H}{\partial e_1} + M_0^2 \frac{\partial G}{\partial e_1}] \tag{6}$$

Using the equilibrium condition of magnetization from equation (4),

$$c_{11} = \frac{\partial}{\partial e_1}[\frac{\partial H}{\partial e_1} + M_0^2 \frac{\partial G}{\partial e_1}] = \frac{\partial^2 H}{\partial e_1^2} + 2M_0(\frac{\partial M_0}{\partial e_1})\frac{\partial G}{\partial e_1} + M_0^2 \frac{\partial^2 G}{\partial e_1^2} \quad (7)$$

The first term in equation (7) $\frac{\partial^2 H}{\partial e_1^2} = c_{11}^0$, is recognized as the elastic coefficient in the absence of magnetization.

Further, from Eq. (5), $2M_0 \frac{\partial M_0}{\partial e_1} = -\frac{2}{b}\frac{\partial G}{\partial e_1}$ leading to

$$\frac{\partial M_0}{\partial e_1} = -\frac{1}{bM_0}\frac{\partial G}{\partial e_1} \quad (8)$$

Using equations (5) and (8) in (6), we obtain,

$$c_{11} = c_{11}^0 - \frac{2}{b}(\frac{\partial G}{\partial e_1})^2 + M_0^2 \frac{\partial^2 G}{\partial e_1^2} \quad (9)$$

When M_0^2 is considered from Eq. (5), it is easily obtained that,

$$c_{11} = c_{11}^0 - \frac{2}{b}(\frac{\partial G}{\partial e_1})^2 + \frac{a}{b}\frac{\partial^2 G}{\partial e_1^2} \quad (10)$$

where second order contributions from the elastic part of the free energy were neglected. Clearly, the temperature dependence of c_{11} originates in the coupling with the equilibrium magnetization, through its proportionality with $a(T)$, expressed in the last term of the Eq. (10). Using the complete form of the free energy, Eq. (1), the six elastic moduli for a single crystal with tetragonal structure are:

$$c_{11} = c_{11}^0 - \frac{2}{b}d_8^2 + \frac{2a}{b}d_1 = c_{11}^0 - \frac{2}{b}d_8^2 + 2d_1 M_0{}^2 \quad (11)$$

Similarly, $c_{44} = \partial^2 \hat{F} / \partial e_2^2 = \frac{\partial^2 H}{\partial e_2^2} + 2M_0(\frac{\partial M_0}{\partial e_2})\frac{\partial G}{\partial e_2} + M_0^2 \frac{\partial^2 G}{\partial e_2^2} = c_{44}^0 + 2d_5 M_0{}^2 \quad (12)$

$$c_{33} = c_{33}^0 - \frac{2}{b}d_7^2 + 2d_4 M_0{}^2 \quad (13)$$

$$c_{66} = c_{66}^0 + 2d_6 M_0^{\ 2} \tag{14}$$

For mixed coefficients, $c_{12} = \frac{\partial^2 H}{\partial e_1 \partial e_2} - \frac{2}{b}(\frac{\partial G}{\partial e_1})(\frac{\partial G}{\partial e_2}) + \frac{a}{b}\frac{\partial^2 G}{\partial e_1 \partial e_2} = c_{12}^0 - \frac{2}{b}d_8^2 + d_2 M_0^{\ 2}$ (15)

Similarly, $c_{13} = c_{13}^0 - \frac{2}{b}d_7 d_8 + d_3 M_0^2$ (16)

Whereas the above calculations yield expressions for the elastic moduli of single crystals, the elastic moduli for isotropic polycrystals can be estimated using the Voigt averaging scheme, which provides an upper limit for the shear and bulk modulus of a polycrystalline solid under the assumption that the stress is uniform everywhere within the sample [28]. The general expressions for the Voigt approximation for the bulk modulus *K* and shear modulus *S* are given by:

$$K = \frac{1}{9}(c_{11} + c_{22} + c_{33}) + \frac{2}{9}(c_{12} + c_{23} + c_{13}) \tag{17}$$

$$S = \frac{1}{15}(c_{11} + c_{22} + c_{33}) - \frac{1}{15}(c_{12} + c_{23} + c_{13}) + \frac{1}{5}(c_{44} + c_{55} + c_{66}) \tag{18}$$

For a tetragonal lattice, the elastic constants satisfy $c_{11}=c_{22}$, $c_{13}=c_{23}$, $c_{44}=c_{55}$, and other elements other than c_{12} and c_{66} are zero. With all elastic moduli c_{ij} having an M^2 dependence, it is clear that the Voigt average for the polycrystalline bulk and shear modulus will display an M^2 dependence as well. Figures 3 and 4 illustrate the model calculations for the polycrystalline moduli c_{44} (= *S*) and c_{11} (= *K* + 4*S*/3), using $c_{ij} = c_{ij}^0 + D_{ij}M^2 + A_{ij}$ where D_{ij} and A_{ij} are constants comprising of the Landau coefficients *b* and d_l, where *l*=1 to 8 obtained from equations (11-16).

Using the equilibrium value of $M^2 = a/b$, the values of the Landau coefficients corresponding to c_{11}, i.e., $D_{11} \sim 0.34$ GPa/emu^2, and $A_{11} \sim -0.29$ GPa are estimated from the slope and intercept of Δc_{11} *vs.* M^2 in the temperature range (10-50) K below T_c as

shown in figure 3(a), where $\Delta c_{11} = c_{11} - c_{11}^0$. The Landau coefficient for c_{44} i.e., D_{44} ~ 0.105 GPa, and A_{44} ~ -0.02 GPa are estimated from the slope of Δc_{44} *vs.* M^2 in the temperature range (10-50) K below T_c as shown in figure 4(a). c_{11}^0 and c_{44}^0 were estimated assuming a temperature-independent (Varshni-like) behavior of the elastic moduli in the absence of magnetization. The Landau coefficients used to model the elastic moduli are tabulated below:

Elastic moduli	Landau coefficients (GPa/emu^2)	Landau coefficients (GPa)
c_{11}	D_{11} = 0.34	A_{11} = -0.29
c_{44}	D_{44} = 0.105	A_{44} = -0.02

As shown in figures 3(b) and 4(b), using the above values of the Landau coefficients, excellent agreement is found between the measured elastic moduli c_{11} and c_{44} below T_c and the Landau model derived for the polycrystalline $Yb_{14}MnSb_{11}$. A quadratic coupling of elastic strain and the spontaneous magnetization, which is the order parameter here, provides an accurate model for the stiffening of the elastic constants in the $Yb_{14}MnSb_{11}$ below T_c. The elastic properties of the rare earth ferromagnet Gd also exhibit similar lattice stiffening below the ferromagnetic ordering temperature (T_c ~ 293.5 K), and the spontaneous magnetic contribution to the Young's modulus in the metal is found to be proportional to the squared magnetization. [29] This may indicate that the $Yb_{14}MnSb_{11}$ behaves more like a rare earth ferromagnet, with no direct overlap of magnetic orbitals, owing to the distance between the magnetic Mn^{2+} ions. Despite the complex nature of ferromagnetism in the $Yb_{14}MnSb_{11}$, the Landau theory of ferromagnetism based on the simple mean-field theory can successfully model the temperature dependence of the elastic moduli near and below the magnetic ordering temperature T_c.

Conclusions

The elastic moduli of the Zintl ferromagnet $Yb_{14}MnSb_{11}$ have been investigated as a function of temperature and magnetic fields, using resonant ultrasound spectroscopy (RUS). The two elastic constants c_{11} and c_{44} for the polycrystalline $Yb_{14}MnSb_{11}$ show unusual stiffening below the ferromagnetic ordering temperature, $T_c \approx 53$ K that corresponds to a 1% increase in the longitudinal and transverse velocities of sound. A similar behavior is observed in the temperature dependence of the resonant frequencies measured for crystallographically unoriented single crystals of $Yb_{14}MnSb_{11}$. The observed stiffening in the $Yb_{14}MnSb_{11}$ below T_c is found to be independent of applied magnetic fields, B=0-2T. A simple model based on Landau theory for ferromagnetism, using a quadratic coupling between the elastic strain and the ferromagnetic order parameter, successfully models the stiffening of the elastic moduli of the $Yb_{14}MnSb_{11}$ below T_c.

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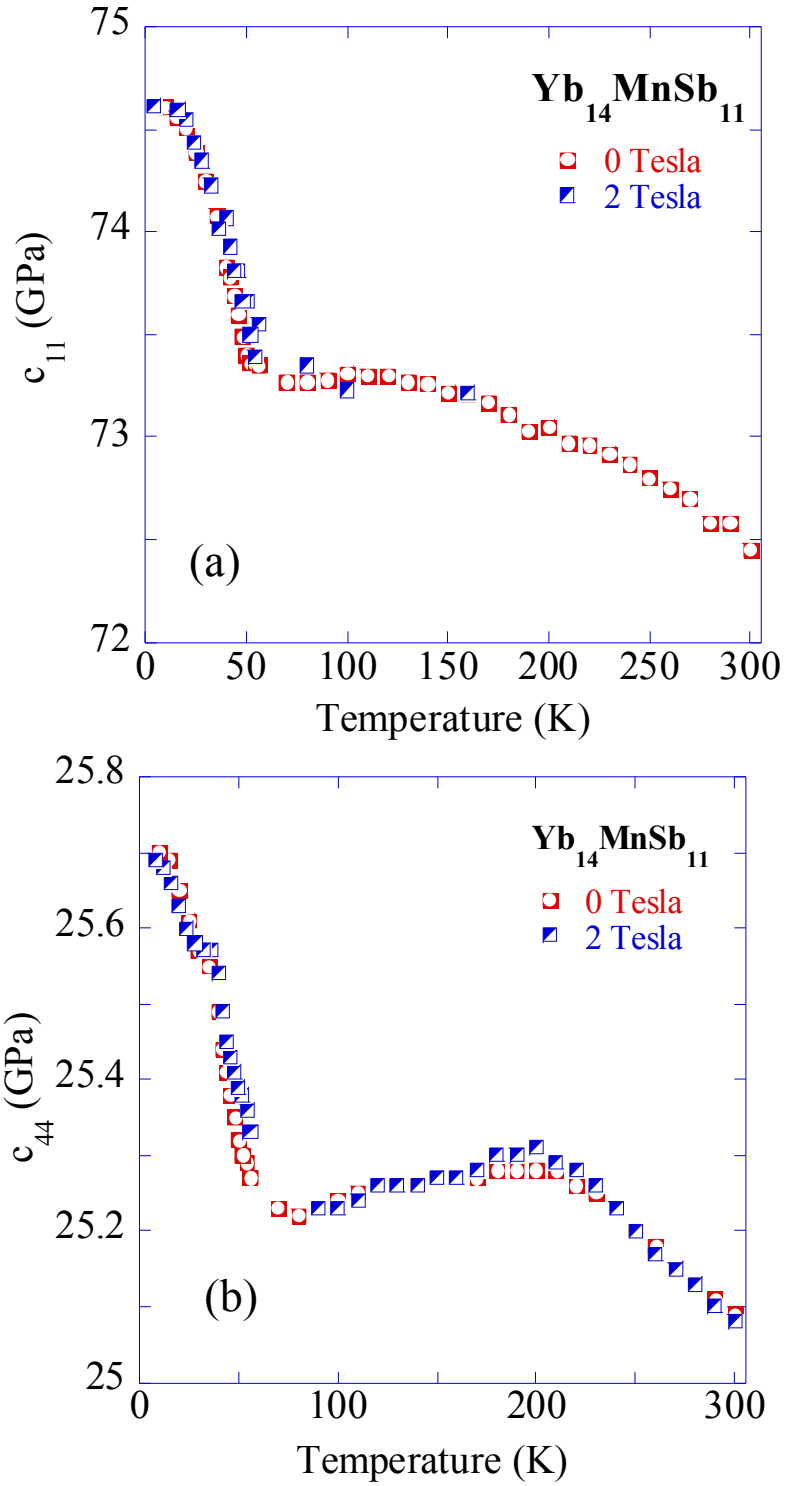

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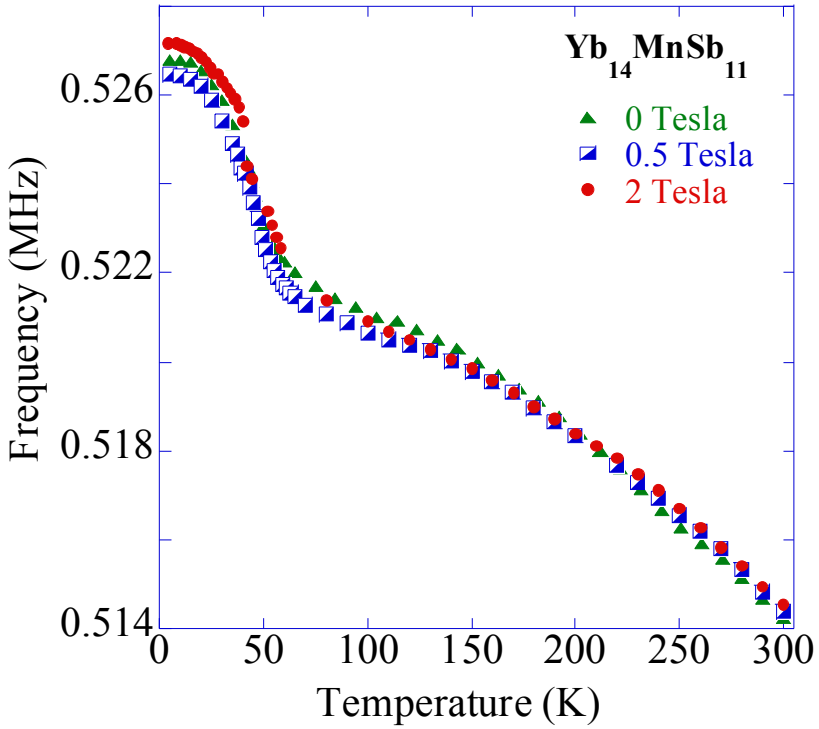

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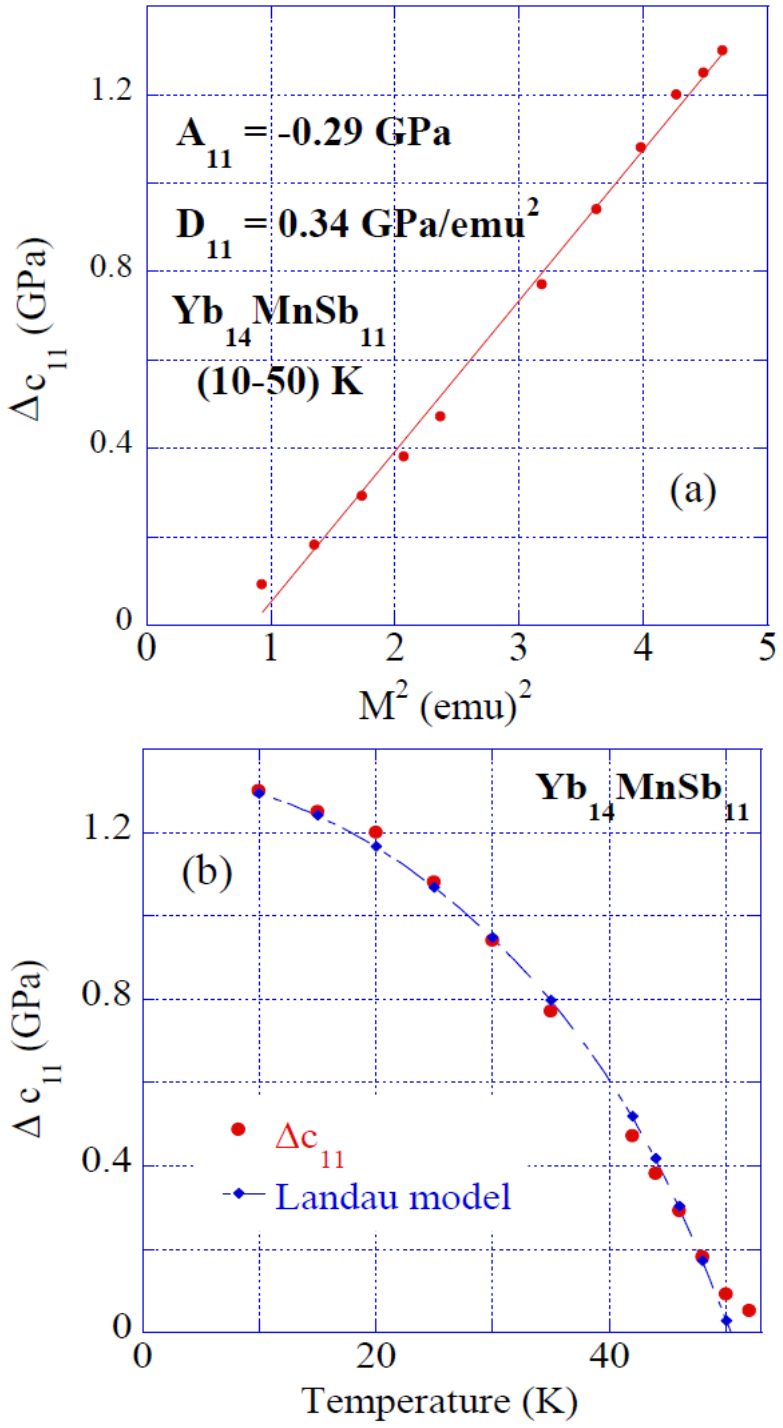

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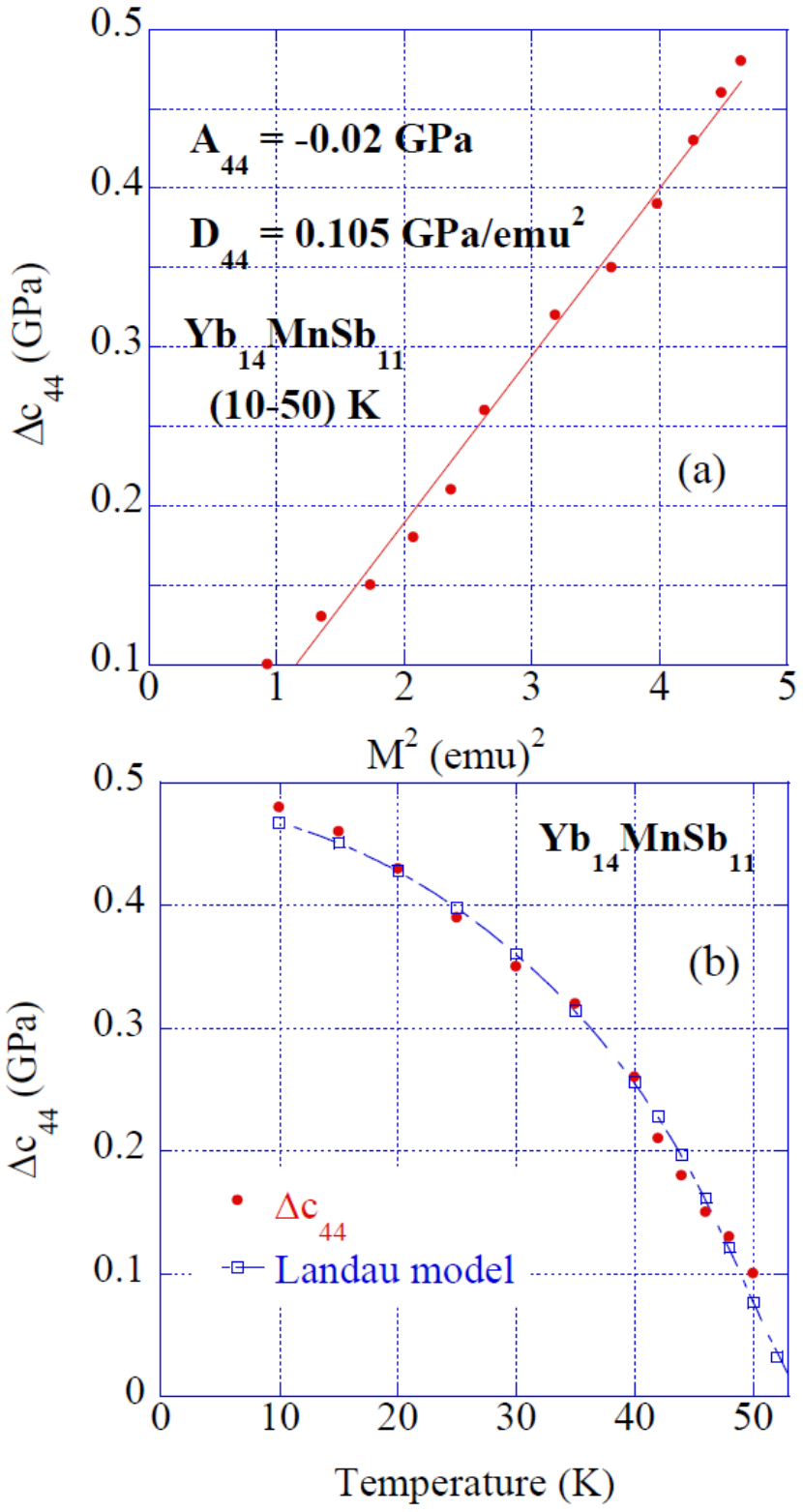

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