

Magnetoelastic anomaly of cubic antiferromagnetic materials

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The magnetoelastic anomaly observed in cubic antiferromagnetic materials is theoretically investigated. The antiferromagnetism of doubly degenerate narrow d bands is described using a two-band Hubbard Hamiltonian and the nesting condition of the band structure. It is shown that the electron redistribution between the antiferromagnetic states introduces a large elastic softening just below the Néel temperature. The theoretical results satisfactorily account for the elastic anomaly observed in chromium and its alloys and, also, in antiferromagnetic Invar.

Among the many physical aspects of antiferromagnetic (AF) materials, the magnetoelastic property poses a very puzzling question. The shear elastic constant c' [$= (c_{11} - c_{12})/2$] shows a large decrease just below the Néel temperature T_N . This elastic anomaly was noted early on Cr,¹ on its alloys,² and on AF Invar such as Fe₆₀Mn₄₀ as shown in Figs. 1–3.³ So far, no satisfactory theory has been forwarded to explain this puzzling behavior.⁴ Although Cr and Fe₆₀Mn₄₀ are all cubic materials, detailed crystal structures are not same. It implies that the anomaly is independent of detailed crystal structures and also of the Invar properties. In this paper, we show that the anomaly originates from the electron distribution between the AF states of the narrow d band structure.

Recently, it was shown that the nature of the elastic softening is closely related to the symmetry of the Fermi level.⁵ Only c' is affected when the Fermi level is of the

e_g type, whereas both c' and c_{44} show anomaly when it is of the t_{2g} type. Since, here, we are mainly concerned with elucidating the physical origin of the elastic anomaly of c' , we assume that the Fermi energy is located on a twofold degenerate e_g band. The case of the threefold degenerate t_{2g} levels will be discussed later. As a model Hamiltonian, we consider the two-band Hubbard Hamiltonian which is given by

$$\begin{aligned}
 H = & \sum_{i,j,\sigma} t_{ij} (c_{i1\sigma}^\dagger c_{j1\sigma} + c_{i2\sigma}^\dagger c_{j2\sigma}) \\
 & + U \sum_i (n_{i1\uparrow} n_{i1\downarrow} + n_{i2\uparrow} n_{i2\downarrow}) \\
 & + U' \sum_{i\sigma\sigma'} n_{i1\sigma} n_{i2\sigma'} - J \sum_{i\sigma} n_{i1\sigma} n_{i2\sigma}
 \end{aligned} \quad (1)$$

where n is the number of electrons, 1 and 2 denote the two degenerate bands, U and U' are the intraband and

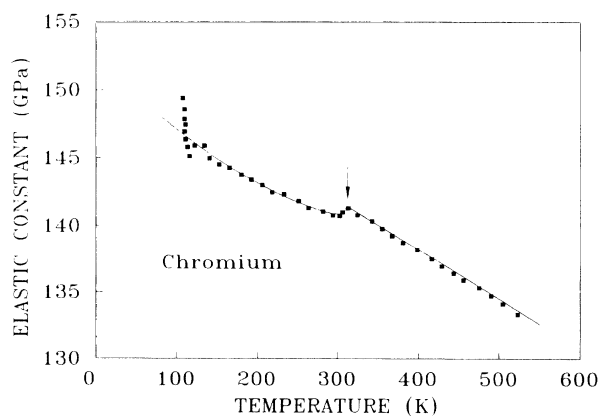


FIG. 1. Elastic constant c' as a function of temperature for Cr (Ref. 1). The full curve represents the theoretical fit. The Néel temperature is indicated by an arrow.

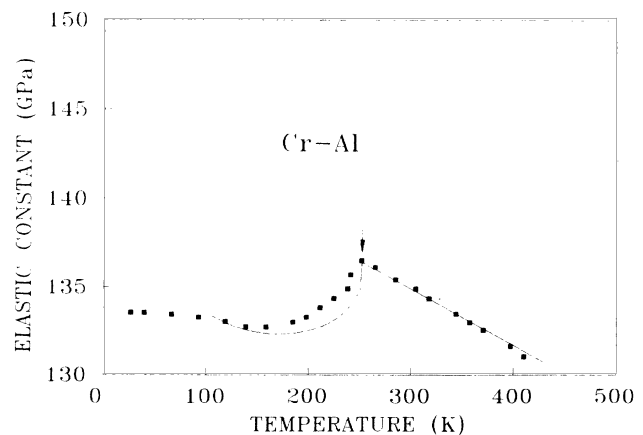


FIG. 2. Elastic constant c' as a function of temperature for Cr-Al alloy (Ref. 2). The full curve represents the theoretical fit.

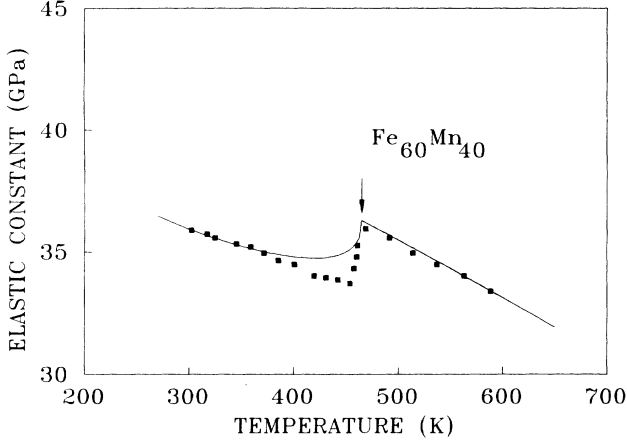


FIG. 3. Elastic constant c' as a function of temperature for antiferromagnetic Invar alloy, $\text{Fe}_{60}\text{Mn}_{40}$ (Ref. 3). The full curve represents the theoretical fit.

the interband on-site Coulomb term, respectively, and J is the interband exchange interaction. The AF eigenvalues of this Hamiltonian have been studied by Ray and Ghatak.⁶ The key ingredient for the existence of the AF state is the formation of a spin-density wave (SDW), whose order parameter is given by $g_{\sigma\sigma'} = \langle c_{1,k,\sigma}^\dagger c_{2,k+Q,\sigma'} \rangle$. For a maximal existence of the antiferromagnetism, we assume $g_{\uparrow\uparrow} = -g_{\downarrow\downarrow}$ and $g_{\uparrow\downarrow} = g_{\downarrow\uparrow} = 0$. We also define a parameter $g = (U' + J)g_{\uparrow\uparrow}$. For formation of the SDW, it is necessary that the band structure satisfies the nesting condition.⁷⁻⁹ With the nesting condition, $\epsilon_1(\mathbf{k}) = -\epsilon_2(\mathbf{k} + \mathbf{Q}) = \epsilon_k$, the energy eigenvalues can be readily obtained following Ref. 6:

$$E_{\pm}^0 = \pm \sqrt{\epsilon_k^2 + g^2 \text{sgn}(\epsilon_k)}. \quad (2)$$

In Ref. 6, and also in other previous calculations,⁷⁻⁹ the energy eigenfunctions were not of primary interest and, thus, were not given explicitly. However, in this calculation, we are interested in the perturbation of the AF energy levels due to the strain and, thus, need the explicit form of the eigenfunctions. The eigenfunctions can also be readily obtained from Eq. (1) using the above eigenvalues and are given by

$$\Psi_{1\pm} = \sin\theta_{k\pm} \Phi_{1k\uparrow} - \cos\theta_{k\pm} \Phi_{2k+Q\uparrow}, \quad (3)$$

$$\Psi_{2\pm} = \sin\theta_{k\pm} \Phi_{1k\downarrow} - \cos\theta_{k\pm} \Phi_{2k+Q\downarrow},$$

where the Bloch wave function $\Phi_{1k\uparrow}$ is given by

$$\Phi_{1k\uparrow} = \sum_i \exp(i\mathbf{k} \cdot \mathbf{R}_i) \phi_1(\mathbf{r} - \mathbf{R}_i) \chi_+. \quad (4)$$

$\Phi_{1k\downarrow}$, $\Phi_{2k+Q\uparrow}$, and $\Phi_{2k+Q\downarrow}$ are also accordingly defined.

$$H_{\text{strain}} = \begin{pmatrix} +E_k^0 + \alpha_+ G_{1+} e & \beta G_2 e & 0 & 0 \\ \beta G_2 e & -E_k^0 + \alpha_- G_{1-} e & 0 & 0 \\ 0 & 0 & +E_k^0 + \alpha_+ G_{1+} e & \beta G_2 e \\ 0 & 0 & \beta G_2 e & -E_k^0 + \alpha_- G_{1-} e \end{pmatrix}. \quad (10)$$

Here ϕ_1 is the basis functions of e_g band with the $3z^2 - r^2$ symmetry and χ_+ is the spinor function. The coefficients $\sin\theta_{k\pm}$ and $\cos\theta_{k\pm}$ are defined by

$$\begin{aligned} \sin\theta_{k\pm} &= R_{\pm} \sqrt{1/(1+R_{\pm}^2)}, \\ \cos\theta_{k\pm} &= \sqrt{1/(1+R_{\pm}^2)}, \end{aligned} \quad (5)$$

and

$$R_{\pm} = \frac{g}{\epsilon_k \mp \sqrt{\epsilon_k^2 + g^2 \text{sgn}(\epsilon_k)}}. \quad (6)$$

Since we are interested in the elastic property of AF materials, we should study the effect of the ultrasonic strain on the AF energy levels. The effect of the strain on a cubic crystal can be represented as $H' = V_{ij} e_{ij}$.⁵ In order to study c' , we consider the corresponding tetragonal strain. When the tetragonal strain is expanded in the irreducible form of the cubic group, the only surviving contribution is given by $e_{31} = e_{xx} + e_{yy} - 2e_{zz} = -2e$.⁶ We now calculate the matrix elements of H' between the AF eigenfunctions. Using the eigenfunctions given above, we arrive at the following expressions after several but straightforward steps:

$$\begin{aligned} \langle \Psi_{1\pm} | V_{31} e_{31} | \Psi_{1\pm} \rangle &= \alpha_{\pm} G_{1\pm} e, \\ \langle \Psi_{1+} | V_{31} e_{31} | \Psi_{1-} \rangle &= \beta G_2 e, \end{aligned} \quad (7)$$

where $\alpha_{\pm} = (\sin^2\theta_{k\pm} - \cos^2\theta_{k\pm})$ and $\beta = (\sin\theta_{k+} \sin\theta_{k-} - \cos\theta_{k+} \cos\theta_{k-})$. $G_{1\pm}$ and G_2 are defined as follows:

$$\begin{aligned} G_{1\pm} &= G + \frac{(\Delta_1 \sin^2\theta_{k\pm} + \Delta_2 \cos^2\theta_{k\pm})}{\alpha_{\pm}}, \\ G_2 &= G + \frac{(\Delta_1 \sin\theta_{k+} \sin\theta_{k-} + \Delta_2 \cos\theta_{k+} \cos\theta_{k-})}{\beta}, \end{aligned} \quad (8)$$

where Δ_1 and Δ_2 are the terms which represent k dependence of the electron-strain interaction and are given by

$$\begin{aligned} \Delta_1(\mathbf{k}) &= \sum_{i \neq j} \exp[-i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)] \\ &\quad \times \langle \phi_1(\mathbf{r} - \mathbf{R}_i) | V_{31}(\mathbf{r}) | \phi_1(\mathbf{r} - \mathbf{R}_j) \rangle, \\ \Delta_2(\mathbf{k}) &= \sum_{i \neq j} \exp[-i(\mathbf{k} + \mathbf{Q}) \cdot (\mathbf{R}_i - \mathbf{R}_j)] \\ &\quad \times \langle \phi_2(\mathbf{r} - \mathbf{R}_i) | V_{31}(\mathbf{r}) | \phi_2(\mathbf{r} - \mathbf{R}_j) \rangle. \end{aligned} \quad (9)$$

The process leading to Eq. (7) can be generalized readily to any type of strain components and, thus, is expected to be useful in studying the strain or structural modulation of the antiferromagnetism.

Now to study the magnetoelastic effect, we add the perturbation to the diagonalized SDW Hamiltonian,

TABLE I. Parameters used to fit the experimental data. D_0 is the maximum density of states at the Fermi level and W is the band width (Ref. 5).

	T_N (K)	D_0 (eV ⁻¹)	W (eV)	$\langle G_{\uparrow}^2 \rangle$ (eV ²)	$\gamma = \langle G_{\downarrow}^2 \rangle / \langle G_{\uparrow}^2 \rangle$	a (Gpa)	b (Gpa/K)
Cr	311	1.50	0.20	5.0	1.25	161.0	-0.044
Cr-Al	252	1.50	0.20	6.6	1.45	155.5	-0.042
Fe ₆₀ Mn ₄₀	465	1.50	0.20	5.0	1.43	54.65	-0.029

This matrix can be diagonalized to yield two doubly degenerate strain-perturbed AF eigenvalues. In order to see the effect of the strain on the AF levels, we expand the eigenvalues up to the second order in strain;

$$E_{\pm} \simeq \pm E_k^0 + \alpha_{\pm} G_{1\pm} e \pm \frac{(\beta G_2 e)^2}{2|E_k^0|}. \quad (11)$$

From Eq. (5) and (6), and from the definitions of α_{\pm} and β , it can be shown easily that $\beta=0$ and $\alpha_+ = -\alpha_-$ above T_N , thus giving $E_{\pm} = \pm E_k^0$, which is the expected paramagnetic result. Below T_N , we observe that the AF ordering enhances the splitting between two e_g levels. The expression for the elastic constant change is given by⁶

$$\begin{aligned} \Delta c' &= \frac{1}{3} \left[\frac{\partial^2 F}{\partial e^2} \right]_{e=0} \\ &= \frac{1}{3} \int d\epsilon D(\epsilon) [f(E) - f(-E)] \left[\frac{\beta^2}{E} \right] \langle G_2^2 \rangle \\ &\quad + \frac{1}{3} \int d\epsilon D(\epsilon) \left[\frac{\partial f(E)}{\partial E} \alpha_+^2 \langle G_{1+}^2 \rangle \right. \\ &\quad \quad \left. - \frac{\partial f(-E)}{\partial E} \alpha_-^2 \langle G_{1-}^2 \rangle \right], \quad (12) \end{aligned}$$

where $\langle G_{1\pm}^2 \rangle$ and $\langle G_2^2 \rangle$ represents the mean values of the deformation potential in the first Brillouin zone. From the first term of the above equation, we observe that the AF ordering introduces a new softening contribution. This additional elastic softening below T_N originates from the redistribution of electrons between the strain-split AF energy bands. The second term of Eq. (12) corresponds to the normal softening term which also exists in the paramagnetic state above T_N . We believe that this AF ordering induced softening clearly explains the magnetoelastic anomaly observed in cubic AF materials as shown in Figs. 1–3. Here, a question may be raised why in ferromagnetic materials, there exists no such elastic anomaly.¹⁰ The answer can be found from the nature of the ferromagnetic transition. Unlike the AF transition which mixes the up and down spins to form a band, the ferromagnetic transition does not mix the spins. Since the perturbing ultrasonic strain does not allow spin-flip transitions, no large additional softening is expected at the Curie temperature.

Although, it is not the main object of this communication to provide an exact quantitative explanation of the magnetoelastic anomaly, nevertheless, we show that the main features of the changes of c' can be fitted satisfactorily using reasonable sets of parameters. Since the first-

principles calculations of the deformation constants are not feasible, we treat them as adjustable parameters to fit the experimental data. In Figs. 1–3, we compared the fitted theoretical results with experimental data. For the fitting process, we assume the lattice contributions to follow a linear relation; $c'_L = a + bT$. Table I shows the sets of parameters to fit the experimental data. The result clearly shows that the present theory satisfactorily explains the magnetoelastic anomaly observed for c' of cubic AF materials. It has been reported that c_{44} of the cubic AF materials do not show any softening effect, but instead shows a slight hardening trends at T_N . Since it is known that c_{44} is not affected, when the Fermi level is of pure e_g nature,⁵ we conjecture that this slight hardening effect may originate from the existence of small t_{2g} level mixing at the Fermi level.

There exists yet another mechanism for the elastic softening.^{11,12} In this mechanism, electrons redistribute between strain-shifted energy pockets which are located at the equivalent \mathbf{k} points of the Brillouin zone. Since it is known that this mechanism is quite general and dominant when the Fermi level is not located at the zone center, we examine, here, the possibility of this mechanism in the AF materials. The AF transition considered above mixes the electron and hole pockets which satisfy the nesting condition and introduces an AF splitting. However, this AF transition does not have any effect on the relative positions of the energy pockets located at the equivalent \mathbf{k} points. Therefore, no new softening contribution is expected from the inter-pocket mechanism below the Néel temperature. The distinct property of the elastic anomaly in AF materials is the sharp decrease of the elastic constant at the Néel temperature, which is only possible when there exists a new softening contribution. Therefore, we conclude that the inter-pocket redistribution cannot be a dominant mechanism for the elastic anomaly in AF materials.

In conclusion, we have studied the magnetoelastic anomaly of the cubic AF materials theoretically. In the process, we obtained the AF energy eigenstates and the strain perturbation of the AF energy levels when the Fermi level is of the e_g type. We believe that this procedure can be easily generalized and will be useful when one is interested in the strain modulation of the AF ordering. Finally, we have shown that the large elastic softening below T_N can be attributed to the electron redistribution between the strain perturbed AF energy states.

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