On the Role of Electronic Polarization in Continuous Structural Transitions

Xiaodong Yang[1], Peter S. Riseborough[* 1], J. L. Smith[2], S. M. Shapiro[3] and J. C. Lashley[2]

[1] Physics Dept. Temple University, Philadelphia PA 19122, USA.
[2] Los Alamos National Lab., Los Alamos, New Mexico 87545, USA.
[3] Brookhaven National Lab., Upton, New York 11973, USA.

Received 29 May 2009, revised 15 Sept 2009, accepted 15 Sept 2007
Published online XXXX

PACS 81.30.Kf, 71.20.Be, 75.40.-s, 77.22.-d

* Corresponding author: e-mail prisebor@temple.edu, Phone: +01 215 204 5655, Fax: +01 214 304 5652.

Martensitic transitions are defined to be diffusionless structural transitions that lower the crystal symmetry and in which the order parameter, usually strain and shuffle, changes discontinuously. It had been proposed that martensitic transitions are driven by the entropy, due to the soft phonons. Anderson and Blount have shown that it is highly improbable that structural transitions are second-order (continuous), and they have suggested that the apparently continuous structural transition in V_3Si is a ferroelectric transition. Recently, a continuous structural martensitic transition has also been identified in crystalline AuZn by Lashley *et al.* Furthermore, pressure measurements on AuZn show that the martensitic temperature can be depressed and suggest the existence of a quantum critical point. The exact nature of the apparently continuous structural transitions is still being debated. However, measurements of magneto-acoustic oscillations in the speed of sound of AuZn indicate that the phonon softening may be driven by the polarization of the conduction electrons. This has been confirmed by recent measurements on AuZn and V_3Si in magnetic fields, which have shown that the transitions are intimately linked to the dielectric response, and are in accord with the predictions of Dieterich and Fulde for V_3Si. The field dependence of the electronic polarization and its role in the transition is discussed.

1 Introduction Martensitic transitions are often defined as diffusionless structural transitions that lower the symmetry, and in which the order parameter has a discontinuity. This definition is generic and is compatible with a group theoretical analysis performed by Anderson and Blount [1] that showed that the transition is usually first order and could only be second order with probability zero. Anderson and Blount then proceeded to argue that the apparently second-order martensitic transition in V_3Si may be an example of a ferroelectric transition. Recently, Lashley *et al.* performed inelastic neutron scattering measurements on the Hume-Rothery alloy AuZn [2] that showed that although the transition was strongly first order in the non-stoichiometric compounds, the hysteresis was greatly reduced, and the order parameter extracted from the satellite intensity was a continuous function of temperature for stoichiometric AuZn. A continuous structural martensitic transition has also been identified in AuCd strain glass by Wang *et al.* [3]. The mechanism responsible for the continuous transitions has not been determined.

We consider the class of materials including AuZn and V_3Si in which the lattice dynamics and structural transition is strongly affected by an applied magnetic field [4, 5]. Due to the low martensitic transition temperature in AuZn, magneto-acoustic oscillations are observable in the speed of sound. The frequency of oscillations are in accord with the frequencies measured in de Haas - van Alphen measurements, and the temperature dependence of the amplitudes are in accord with the Lifshitz-Kosevich formulae [4]. A theoretical framework has been created [6] that describes the effect of the field on the lattice dynamics in which the field dependence originates from the dielectric constant. It is believed that the entropy produced by the soft phonons [7,8] is responsible for driving the martensitic transitions. The weak field dependence of the martensitic transition temperature in AuZn is in strong contrast with the strong field dependence in V_3Si [5], and the difference has been related to the difference in the dimensionality of these two materials [6,9,10].

2 Lattice Dynamics The phonon frequencies $\omega_\alpha(\underline{q})$ and the polarization vectors $\underline{\epsilon}_\alpha(\underline{q})$ are determined from the eigenvalue equation

$$\frac{M}{N}\,\omega^2_\alpha(\underline{q})\,\underline{\epsilon}_\alpha(\underline{q}) = \sum_{\underline{Q}} (\,\underline{q}+\underline{Q}\,)\,\Theta(\underline{q}+\underline{Q})\,(\,\underline{q}+\underline{Q}\,)\cdot\underline{\epsilon}_\alpha(\underline{q}) - \sum_{\underline{Q}} \underline{Q}\,\Theta(\underline{Q})\,(\,\underline{Q}\cdot\underline{\epsilon}_\alpha(\underline{q})\,) \qquad (1)$$

where the sum over $\underline{Q}$ is a sum over reciprocal lattice vectors and where the Fourier Transform of the pair-potential $\Theta(\underline{k})$ is approximated by the screened Coulomb interaction

$$\Theta(\underline{k}) \;=\; \frac{1}{V}\left(\frac{4\,\pi\,Z^2\,e^2}{k^2\,\epsilon(\underline{k})}\right)\widetilde{V}_0(k)^2 \qquad (2)$$

and $\widetilde{V}_0(k)$ is a dimensionless oscillatory function of k that only depends on the core radius [11]. Therefore, the field-dependence of the phonon frequencies originates from the dielectric constant and can be expressed in terms of the Lindhard function. The low-temperature limit of the Lindhard function can be expressed as a sum over the occupied Landau levels n

$$\chi(\underline{Q})_H = -\frac{1}{2\pi^2\hbar\omega_c Q_z r_c^4}\sum_{n,m,\sigma}\,|\,F_{n,m}(Q_\perp)\,|^2 \times \ln\left|\frac{2m_e(n-m)\omega_c/\hbar - Q_z^2 - 2Q_z k_{F,n\sigma}}{2m_e(n-m)\omega_c/\hbar - Q_z^2 + 2Q_z k_{F,n\sigma}}\right| \qquad (3)$$

where $k_{F,n\sigma}$ is the Fermi wave vector of the n-th spin split occupied Landau level

$$k_{F,n\sigma} \;=\; \sqrt{\frac{2m_e}{\hbar^2}\left[\,\epsilon_F - \left(n+\frac{1-\sigma}{2}\right)\hbar\omega_c\,\right]} \qquad (4)$$

and ω_c is Larmour frequency. The form factor $F_{n,m}(Q_\perp)$ is given by

$$F_{n,m}(Q_\perp) = \left(\frac{n!}{m!}\right)^{\frac{1}{2}}\left(\frac{r_c\,(\,i\,Q_x\,-\,Q_y\,)}{\sqrt{2}}\right)^{m-n} \times\,\exp\left[\,-\,\frac{r_c^2\,Q_\perp^2}{4}\,\right]L_n^{m-n}\left(\frac{r_c^2\,Q_\perp^2}{2}\right) \qquad (5)$$

for $m > n$, where r_c is the radii of the Landau orbits and where $L^n_m(x)$ represents the associated Laguerre functions. The field-dependence at an arbitrary $\underline{Q}$ value is shown in fig(1). The singularities occur for fields where $\underline{Q}$ connect the surfaces of Landau tubes (n,m). The points of intersection are shown graphically in fig(2). Since the form factors are rapidly oscillating functions of (n,m) and since they satisfy a sum rule, the oscillations in the phonon frequencies are dominated by $\underline{Q}$ vectors that are either parallel to the applied field or $\underline{Q} = 0$. The oscillations in the sound velocity of AuZn were identified with $Q = 0$. Since the transition temperature is relatively low, the application of pressure can significantly reduce the transition temperature [2]. The transition temperature pressure relation is expected to deviate from linearity at low tempera-

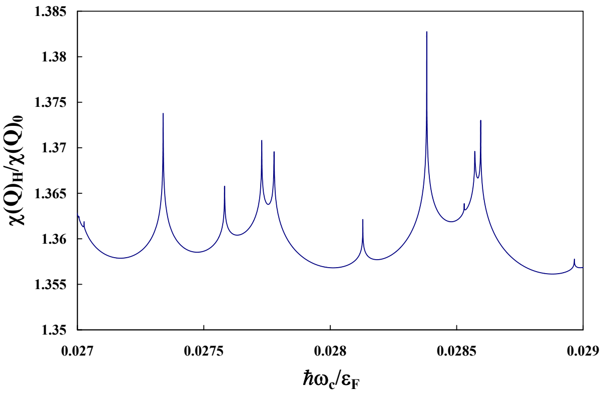

Figure 1 The schematic field-dependence of the (normalized) density-density response function, $\chi(\underline{Q})_H$ on $\hbar\omega_c/\epsilon_F$, for a general direction of Q such as $Q_\perp/k_F = Q_z/k_F = 1/3$.

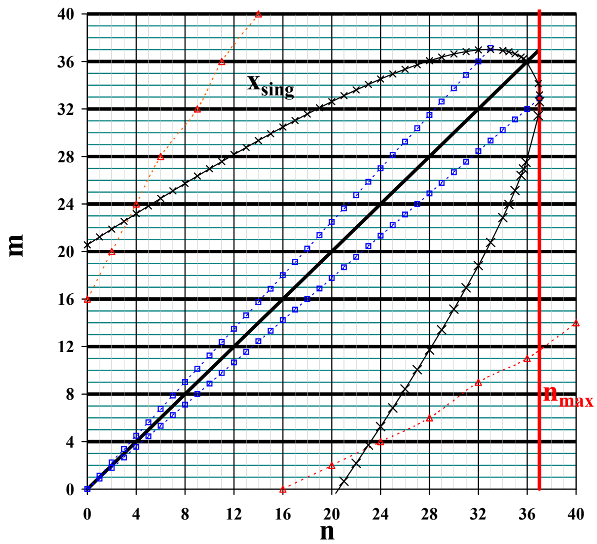

Figure 2 The (n,m) phase space for Landau levels. The maximum value of n (marked by a solid red line) is determined by the ratio of the Fermi-energy to $\hbar\omega_c$. A symmetric pair of singularities occur for fields where $\underline{Q}$ connects the surfaces of two Landau tubes. These are represented by the intersections of the parabola (black crosses) with lattice points. The singularities add inside the region marked by the open blue squares and cancel outside this region.

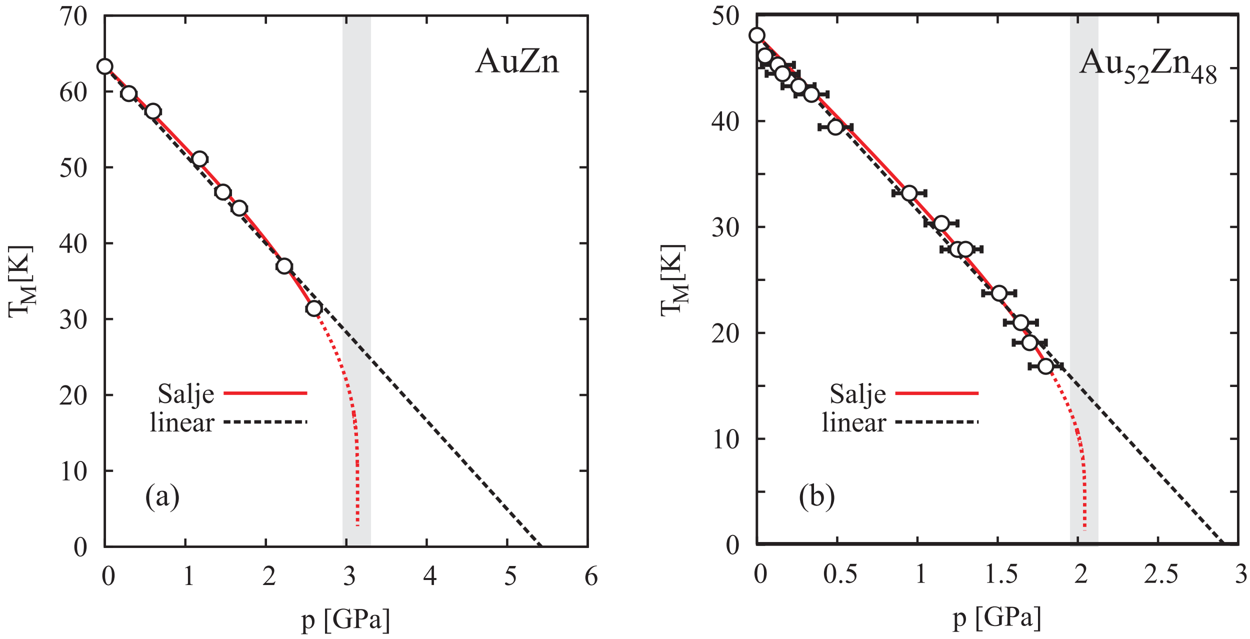

Figure 3 The P-T phase diagram for AuZn and $Au_{52}Zn_{48}$, together with linear (black) and non-linear (red) extrapolations of the phase boundaries. The data are taken from reference [2]. The non-linear extrapolations are based on the theory of reference [12].

tures and asymptotically approach zero with infinite slope due to Nernst's law [12]. However, the approach to a Quantum Critical Point is expected to be preempted. Following Landau and Lifshitz's [13] discussion of systems where the transition temperature depends on pressure (for which volume terms must be included the Gibbs free-energy functional), it is expected that the line of second-order transitions will be continued by a line of first-order transitions when the magnitude of the slope of the curve exceeds a critical value. For values of P greater than the critical value determined in terms of the compressibility, the state with continuously broken symmetry still exists but only as an excited state.

3 Conclusions The structural and lattice dynamical properties of materials such as AuZn and V_3Si that exhibit continuous structural transitions which are strongly influenced by the effects of magnetic fields. This has been explained in terms of the dielectric response. For quasi-low-dimensional materials such as V_3Si, the enhanced dielectric response is expected in terms of Fermi-surface nesting. For good three-dimensional materials such as AuZn, where Fermi-surface nesting also occurs, the enhanced dielectric response could be considered as support for the hypothesis that the continuous martensitic transition is a screened ferroelectric transition [1].

Acknowledgements The work at Temple University was supported by the U.S. Department of Energy Office of Basic Energy Sciences by DEFG02-01ER45872. The work at Los Alamos Nat. Lab. and Brookhaven Nat. Lab. was performed under the auspices of the U.S. Department of Energy.

References

[1] P. W. Anderson and E. I. Blount, Phys. Rev. Lett. **14**, 217 (1965).
[2] J. C. Lashley, S. M. Shapiro, B. L. Winn, C. P. Opeil, M. E. Manley, A. Alatas, W. Radcliffe, T. Park, R. A. Fisher, B. Mihaila, P. S. Riseborough, E. K. H. Salje and J. L. Smith, Phys. Rev. Lett. **101**, 135703 (2008).
[3] Y. Wang, X. Ren and K. Otsuka, Phys. Rev. Lett. **97**, 225703 (2006).
[4] J. C. Lashley, R. K. Schulze, B. Mihaila, W. L. Hults, J. C. Cooley, J. L. Smith, P. S. Riseborough, C. P. Opeil, R. A. Fisher, O. Svitelskiy, A. Suslov and T. R. Finlayson, Phys. Rev. B, **75**, 205119 (2007).
[5] P. A. Goddard, A. V. Silhanek, and J. C. Lashley, *in preparation* (2009).
[6] X-D. Yang, P. S. Riseborough, K. A. Modic, R. A. Fisher, C. P. Opeil, T. R. Finlayson, J. C. Cooley, J. L. Smith, P. A. Goddard and J. C. Lashley, Phil. Mag. **89**, 2083 (2009).
[7] Clarence Zener, Phys. Rev. **71**, 846 (1947).
[8] J. Friedel, J. de Physique **36**, L69 (1974).
[9] J. Labbe and J. Friedel, J. Physique **27**, 153, 303 (1966).
[10] W. Dieterich and P. Fulde, Z. Physik **248**, 154 (1971).
[11] V. Heine and D. Weaire, Phys. Rev., **152**, 603 (1966).
[12] S. A. Hayward and E. K. H. Salje, J. Phys.: Cond. Matt., **10**, 1421 (1998),
[13] L. D. Landau and E. M. Lifshitz, *Statistical Mechanics*, Pergamon Press, Oxford 2001.