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Direct evidence of the magnetoelastic interaction in Ni_2MnGa magnetic shape memory system

V. Recarte,[1,a)] J. I. Pérez-Landazábal,[1] V. Sánchez-Alarcos,[1] E. Cesari,[2] M. Jiménez-Ruiz,[3] K. Schmalzl,[4] and V. A. Chernenko[5,6]

[1]*Departamento de Física, Universidad Pública de Navarra, Campus de Arrosadía 31006 Pamplona, Spain*
[2]*Departament de Física, Universitat de les Illes Balears, Ctra. de Valldemossa, km 7.5, E-07122 Palma de Mallorca, Spain*
[3]*Institut Laue Langevin (ILL), 6 Rue Jules Horowitz, BP 156, 38042 Grenoble, France*
[4]*Juelich Centre for Neutron Science JCNS, Forschungszentrum Juelich GmbH, Outstation at ILL, BP 156, 38042 Grenoble, France*
[5]*BCMaterials & Universidad del País Vasco, 48080 Bilbao, Spain*
[6]*IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Spain*

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The martensitic transformation in stoichiometric Ni_2MnGa alloys is preceded by a weakly first order transformation from a high temperature cubic phase to a near-cubic modulated intermediate phase related to the presence of a soft phonon mode. This transformation has been proposed to appear as a consequence of the magnetoelastic coupling. Inelastic neutron scattering experiment performed under external magnetic field shows a temperature shift of the characteristic energy dip at $\zeta \approx 0.33$. Furthermore, an enhancement of the long-wavelength limit (C') of this branch with the applied magnetic field has been observed. Both results evidence a strong magnetoelastic interaction at the intermediate transition. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4807737]

Ni-Mn based magnetic shape memory alloys have received great attention during the last years due to their novel functional properties such as the large magnetic field induced strain[1,2] and magnetocaloric effect.[3] These properties have the origin on the martensitic transformation (MT) from a cubic $L2_1$-ordered parent phase (*P*-phase, $Fm\bar{3}m$ space group) to a low-temperature low symmetry martensitic phase. The origin of this transformation is ascribed to Jahn-Teller distortions and Fermi-surface nesting at electronic structure level.[4] Prior to the MT, the parent phase commonly undergoes precursor effects, as the softening of the low-lying transverse TA_2-phonon branch,[5–9] linked to the weak restoring forces that arise in some specific crystallographic directions as a consequence of the dynamical instability of the cubic structure.

In some Ni-Mn-Ga alloys close to the Ni_2MnGa stoichiometry, the MT is also preceded by a weakly first order transformation from the *P*-phase into a near-cubic intermediate phase (*I*-phase) with a six-layered lattice modulation.[10,11] Several precursor phenomena occur prior to the *P-I* transformation during cooling, among them the temperature dependent dip at a wave vector $\zeta \approx 0.33$ inside the Brillouin zone (intracell distortion)[5,12,13] and the softening of the long-wavelength limit of the transverse-acoustic TA_2-phonon branch at $\zeta = 0$ (that is, the softening of the C' shear modulus).[14–16] On the other hand, an up-turn of both the phonon frequency of the soft mode and C' is observed below the *P-I* transformation temperature, T_I. The intermediate transformation has been shown both experimentally and theoretically to be a result of the electron-phonon coupling and Fermi surface nesting enforced by a strong magneto-elastic interaction.[17–24] The key role of magnetism in this transformation, and in particular the requirement of a well-developed ferromagnetic order for the transition to take place, accounts for the fact that the intermediate transition has just been observed on Ni-Mn-Ga alloys whose MT occurs far below the Curie temperature, as well as for the lack of intermediate phase in the metamagnetic Ni-Mn-X (X = In, Sn, Sb). In keeping with this, the first order character of the *P-I* transformation has been explained by a Landau model, where the magnetization and the anomalous phonon amplitude are coupled.[22]

Experimental studies on the influence of magnetic field on the intermediate transition evoked some controversy. First magnetization studies suggested that the transition is suppressed at high values of the applied magnetic field,[25] while resistivity measurements did not reveal any influence of the magnetic field.[26] Furthermore, most of the works reported that low magnetic fields shift the transition to lower temperatures[27–31] but for large applied magnetic fields a positive shift of the temperature has been detected.[31] However, the influence of the magnetic field on the dynamics of the soft phonon mode, which would indeed provide a direct evidence of the magnetoelastic coupling, has still not been studied. In this respect, we report an inelastic neutron scattering study of the magnetic field influence on the behavior of the TA_2-phonon branch in Ni_2MnGa.

A stoichiometric Ni_2MnGa single crystalline alloy was prepared by the Bridgman method. The Curie temperature, $T_C = 380$ K, martensitic transformation temperature, $T_M = 175$ K, and intermediate transformation temperature, $T_I \approx 240$ K, were determined. A rectangular parallelepiped of $6.7 \times 4.8 \times 11.5$ mm^3 with faces parallel to (110), $(1\bar{1}0)$, and (001) planes was prepared for the neutron experiment. The TA_2-phonon branch was measured in the 180 K–260 K temperature range, and without applied magnetic field and under a magnetic field of 60 KOe applied in the (001) direction. Neutron scattering experiments were carried out on the cold

a)Author to whom correspondence should be addressed. Electronic mail: recarte@unavarra.es. Tel.: +34 948 169578, Fax: +34 948 169565.

neutron triple axis spectrometer IN12 at the Institute Laue-Langevin. The optimal conditions for accessing the TA_2-phonon with a good energy resolution were found to be those employing the PG (002) monochromator and PG (002) analyzer, using horizontal collimations of 30 in. and a constant final wave vector $k_f = 1.97\ Å^{-1}$. A PG filter was placed between the sample and the analyser. For the optical branch for a value of $\xi = 0.3$ and higher, a smaller wave vector was used in order to avoid the presence of spurions on the inelastic spectra. For such measurements, a constant final wave vector of $k_f = 1.85\ Å^{-1}$ was used.

Fig. 1 shows the TA_2-phonon branch along the [110] direction with polarization in the $[1\bar{1}0]$ direction at T = 240 K (close to T_I). Due to the limitation in energy transfer, it is not possible to reach the limit of the Brillouin zone but the main feature of this soft phonon branch, the dip at $\zeta \approx 0.33$, appears clearly defined. This wiggle in the TA_2-phonon branch, which also appears at temperatures above the Curie point,[5] becomes more pronounced as the temperature decreases, being the degree of softening enhanced by the magnetic order. The higher degree of softening is observed at T_I, the temperature of the intermediate transition in which the freezing of instabilities develops in a new phase.[5,12] To determine the effect of an external magnetic field on the vibrational response of the alloy, the behavior of the softening of the TA_2 phonon branch under a 60 KOe external magnetic field perpendicular to the scattering plane has been analyzed. Measurements under H = 0 Oe and H = 60 KOe for ζ values around the deep in the temperature range between 210 K and 260 K are shown in Figure 2. Focusing on the zero field results, the minimum value of the energy $E \approx 0.8$ meV is observed at 240 K, close to intermediate transformation temperature, $T_I \approx 240$ K. On the other hand, the minimum value of energy under magnetic field ($E \approx 0.85$ meV) was detected at 250 K. For temperatures between 210 K and 240 K, the magnetic field stiffens the soft mode, changing the effect at higher temperatures. A clear influence of the magnetic field on the position of the phonon minimum, $\zeta \approx 0.33$, cannot be inferred from experimental results, although previous calculations proposed that the phonon softening wave vector could be changed by the application of an external magnetic field.[18]

In accordance with the soft mode theory, the square of the energy at the dip, $(\hbar\omega)^2$, would decrease linearly with the

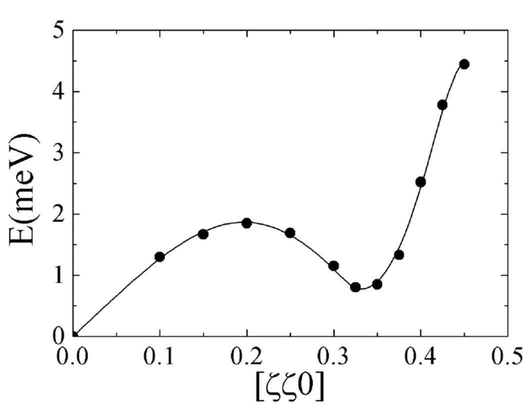

FIG. 1. TA_2 phonon branch at T = 240 K showing the minimum value of the energy at the dip, $\zeta \approx 0.33$.

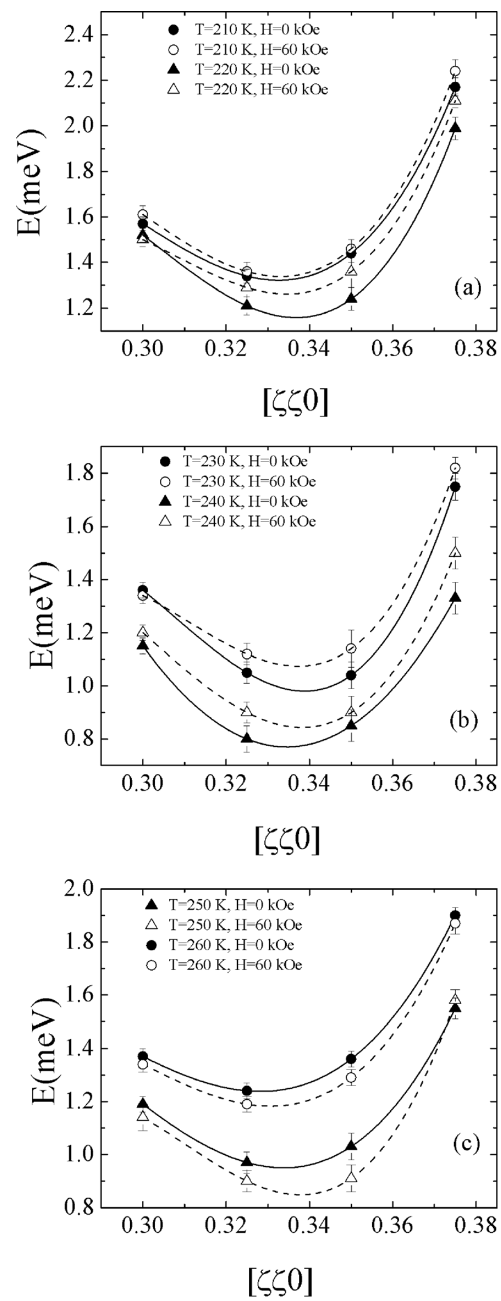

FIG. 2. TA_2 phonon branch for ζ values around the dip for H = 0 and 60 KOe: (a) at 210 K and 220 K; (b) at 230 K and 240 K; (c) at 250 K and 260 K. Lines are guidelines for the eyes.

decreasing temperature till the lattice becomes dynamically unstable and a second order transition took place at T_o, where $\hbar\omega^2 = 0$. In the case of Ni_2MnGa, in turn, the magnetoelastic coupling enables the existence of the intermediate phase transition at $T_I > T_o$ before the phonon softening is completed.[22] A characteristic of the intermediate phase is the stiffening of the TA_2 phonon branch with decreasing temperature, showing $(\hbar\omega)^2$ a minimum at T_I.[5,12] The TA_2 phonon squared energy E^2 versus temperature (under H = 0 and H = 60 KOe) for the closest values to the minimum ($\zeta = 0.325$ and $\zeta = 0.350$) are shown in Figure 3. Despite both curves show similar dependence on temperature, a clear shift of the temperature of the minimum, $\Delta T \approx 4$ K, is observed as a consequence of the application of the magnetic field. It demonstrates that, due to the spin-phonon

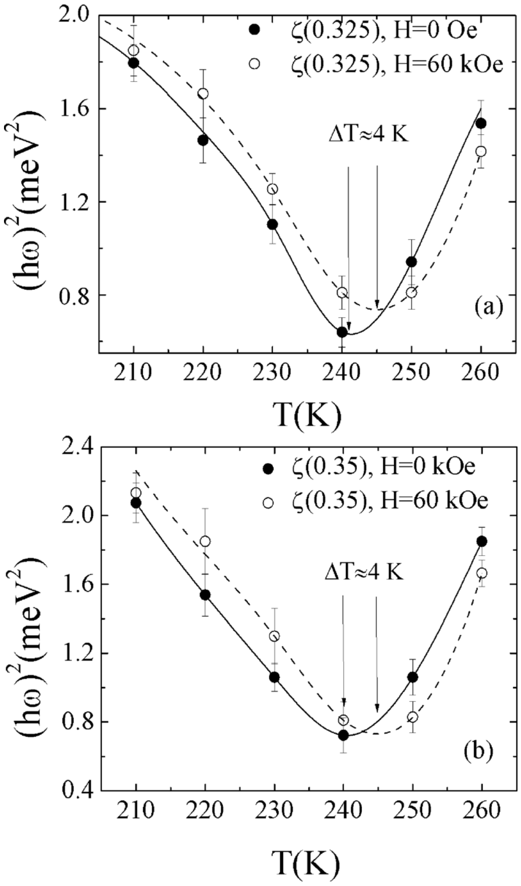

FIG. 3. Square of the energy $(\hbar\ \omega)^2$ versus temperature for the $[\zeta\zeta 0]$ TA_2 branch at: (a) $\zeta = 0.325$ and (b) $\zeta = 0.325$ without applied magnetic field and under a magnetic field of 60 KOe. The temperature of the minimum increases 4 K due to the application of the magnetic field. Lines are guidelines for the eyes.

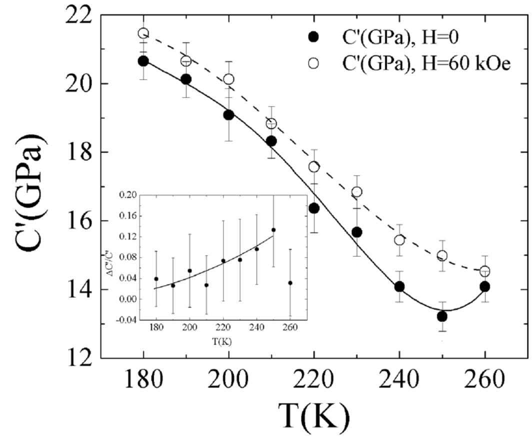

FIG. 4. Elastic constant C' versus temperature without applied magnetic field and under a magnetic field of 60 KOe. The inset shows the normalized increased of C', $\Delta C'/C' = \left(C'(\mathrm{H}) - C'(0)\right)/C'(0)$.

interaction, the external magnetic field stabilizes the intermediate phase against the cubic structure. The variation of the position of the E^2 minimum is in good agreement with the dependence of the temperature of the intermediate transition under high magnetic fields determined by DC resistance measurements, $dT_I/dH = 0.06$ K/kOe.[31] The observed shift is also in agreement with recent *ab initio* calculations carried out by constraining the magnetic moment to a predefined value (as a way to account for magnetic field effects), which show that the instability at $\zeta \approx 0.33$ increases with increasing magnetization destabilizing the cubic $L2_1$ parent phase.[32]

In this sense, the stabilization of the intermediate phase by magnetism has been recently shown to be also promoted by variations in the magnetic exchange coupling linked to long-range atomic order variations. The atomic order of the $L2_1$ cubic phase influences the magnetic exchange coupling as the magnetic interactions depend on the Mn-Mn distance. The dependence of magnetic and structural transition temperatures on the next-nearest-neighbors atomic degree of order η has been quantified for Ni-Mn-Ga alloys[33] and explained in terms of the effect of the magnetic exchange coupling variations on the free energy difference between the transforming phases.[34] In particular, it has been demonstrated that the increase of $L2_1$ order parameter shifts T_I to higher temperatures due to the increase of magnetic moment.[35] It is therefore interesting to note that, through the magnetic coupling enhancement, an external magnetic field acts in the same way as atomic order variations.

Another feature of the magnetoelastic coupling is the change in the elastic constants under applied magnetic field. In our case, the C' elastic constant can be determined from the initial slope of the TA_2-phonon branch. To make estimation, the lowest measured point $\zeta \approx 0.1$ has been taken, being the error in the slope determined by the error in the value. Figure 4 shows the temperature dependence of C' with and without applied magnetic field. In both cases, C' increases on cooling below T_I, in agreement with previous studies on the elastic response of the intermediate phase.[13] The elastic constant increases with the magnetic field in the whole temperature range between T_I and T_M, i.e., the temperature range of the existence of the intermediate phase. In addition, the position of the minimum, which is close to T_I in the zero field case, is shifted in agreement to the previously observed shift of T_I *(H)*. The inset in Figure 4 shows the relative change in the elastic constant $\Delta C'/C' = (C'(H) - C'(0))/C'(0)$ as a function of temperature. The maximum value $\Delta C'/C' \approx 0.14$ takes place at T_I, decreasing to zero when the temperature moves away from T_I. This large relative change, which is one order of magnitude higher than the change observed at temperatures far above T_I $(\Delta C'/C' \approx 0.015)$,[36] evidences the strong magnetoelastic interaction present at the intermediate transition.

In summary, the influence of the magnetic field on the TA_2-phonon branch in the temperature range where the intermediate transition takes places has been measured in a Ni_2MnGa alloy. The observed behavior, the shift of the characteristic deep at $\zeta \approx 0.33$ and the enhancement of the long-wavelength limit (C') of this low laying branch with the applied magnetic field evidences the strong magnetoelastic interaction at the intermediate transition and reflects the outstanding role of the magnetism at this transition.

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[1]K. Ullakko, J. K. Huang, C. Kantner, R. C. O'Handley, and V. V. Kokorin, Appl. Phys. Lett. **69**, 1966 (1996).
[2]R. Kainuma, Y. Imano, W. Ito, Y. Sutou, H. Morito, S. Okamoto, O. Kitakami, K. Oikawa, A. Fujita, T. Kamomata, and K. Ishida, Nature **439**, 957 (2006).
[3]A. Planes, L. Mañosa, and M. Acet, J. Phys.: Condens. Matter **21**, 233201 (2009).
[4]P. J. Brown, A. Y. Bargawi, J. Crangle, K.-U. Neumann, and K. R. A. Ziebeck, J. Phys.: Condens. Matter **11**, 4715 (1999).
[5]A. Zheludev, S. M. Shapiro, P. Wochner, A. Schwartz, M. Wall, and L. Tanner, Phys. Rev. B **51**, 11310 (1995).
[6]X. Moya, L. Mañosa, A. Planes, T. Krenke, M. Acet, O. Garlea, T. A. Lograsso, D. L. Schlagel, and J. L. Zarestky, Phys. Rev. B **73**, 064303 (2006).
[7]T. Mehaddene, J. Neuhaus, W. Petry, K. Hradil, P. Bourges, and H. Hiess, Phys. Rev. B **78**, 104110 (2008).
[8]X. Moya, D. Gonzalez-Alonso, L. Mañosa, A. Planes, O. Garlea, T. A. Lograsso, D. L. Schlagel, J. L. Zarestky, S. Aksoy, and M. Acet, Phys. Rev. B **79**, 214118 (2009).
[9]J. I. Pérez-Landazábal, V. Recarte, V. Sánchez-Alarcos, J. A. Rodríguez-Velamazán, M. Jiménez-Ruiz, P. Link, E. Cesari, and Y. I. Chumlyakov, Phys. Rev. B **80**, 144301 (2009).
[10]V. V. Kokorin, V. A. Chernenko, E. Cesari, J. Pons, and C. Segui, J. Phys.: Condens. Matter **8**, 6457 (1996).
[11]V. A. Chernenko, J. Pons, C. Seguí, and E. Cesari, Acta Mater. **50**, 53 (2002).
[12]A. Zheludev, S. M. Shapiro, P. Wochner, and L. Tanner, Phys. Rev. B **54**, 15045 (1996).
[13]U. Stuhr, P. Vorderwisch, V. Kokorin, and P.-A. Lindgård, Phys. Rev. B **56**, 14360 (1997).
[14]L. Mañosa, A. González-Comas, E. Obradó, A. Planes, V. A. Chernenko, V. V. Kokorin, and E. Cesari, Phys. Rev. B **55**, 11068 (1997).
[15]T. E. Stenger and J. Trivisonno, Phys. Rev. B **57**, 2735 (1998).
[16]J. I. Pérez-Landazábal, V. Sánchez-Alarcos, C. Gómez-Polo, V. Recarte, and V. A. Chernenko, Phys. Rev. B **76**, 092101 (2007).
[17]O. I. Velikokhatnyi and I. I. Naumov, Phys. Solid State **41**, 617 (1999).
[18]Y. Lee, J. Y. Rhee, and B. N. Harmon, Phys. Rev. B **66**, 054424 (2002).
[19]C. Bungaro, K. Rabe, and A. D. Corso, Phys. Rev. B **68**, 134104 (2003).
[20]C. Opeil, B. Mihaila, R. K. Schulze, L. Mañosa, A. Planes, W. L. Hults, R. A. Fisher, P. S. Riseborough, P. B. Littlewood, J. L. Smith, and J. C. Lashley, Phys. Rev. Lett. **100**, 165703 (2008).
[21]S. W. D'Souza, A. Rai, J. Nayak, M. Maniraj, R. S. Dhaka, S. R. Barman, D. L. Schlagel, T. A. Lograsso, and A. Chakrabarti, Phys. Rev. B **85**, 085123 (2012).
[22]A. Planes, E. Obradó, A. González-Comas, and Ll. Mañosa, Phys. Rev. Lett. **79**, 3926 (1997).
[23]T. Castán, E. Vives, and P.-A. Lindgård, Phys. Rev. B **60**, 7071 (1999).
[24]M. A. Uijttewaal, T. Hickel, J. Neugebauer, M. E. Gruner, and P. Entel, Phys. Rev. Lett. **102**, 035702 (2009).
[25]F. Zuo, X. Su, and K. H. Wu, Phys. Rev. B **58**, 11127 (1998).
[26]J. Kim, F. Inaba, T. Fukuda, and T. Kakeshita, Acta Mater. **54**, 493 (2006).
[27]E. Obradó, A. González-Comas, L. Mañosa, and A. Planes, J. Appl. Phys. **83**, 7300 (1998).
[28]W. H. Wang, J. L. Chen, S. X. Gao, G. H. Wu, Z. Wang, Y. F. Zheng, L. C. Zhao, and W. S. Zhan, J. Phys.: Condens. Matter **13**, 2607 (2001).
[29]Y. T. Cui, J. L. Chen, G. D. Liu, G. H. Wu, and W. L. Wang, J. Phys.: Condens. Matter **16**, 3061 (2004).
[30]B. Ludwig, C. Strothkaemper, U. Klemradt, X. Moya, Ll. Mañosa, E. Vives, and A. Planes, Phys. Rev. B **80**, 144102 (2009).
[31]J. M. Barandiaran, V. A. Chernenko, P. Lazpita, J. Gutiérrez, I. Orue, J. Feuchtwanger, and S. Besseghini, Appl. Phys. Lett. **94**, 051909 (2009).
[32]M. E. Gruner, W. A. Adeagbo, A. T. Zayak, A. Hucht, S. Buschmann, and P. Entel, Eur. Phys. J. Spec. Top. **158**, 193 (2008).
[33]V. Sánchez-Alarcos, J. I. Pérez-Landazábal, V. Recarte, J. A. Rodríguez-Velamazán, and V. A. Chernenko, J. Phys.: Condens. Matter **22**, 166001 (2010).
[34]V. Sánchez-Alarcos, V. Recarte, J. I. Pérez-Landazábal, C. Gómez-Polo, and J. A. Rodríguez-Velamazán, Acta Mater. **60**, 459 (2012).
[35]V. Sánchez-Alarcos, J. I. Pérez-Landazábal, and V. Recarte, Mater. Sci. Forum **684**, 85 (2011).
[36]A. González-Comas, E. Obradó, L. Mañosa, A. Planes, V. A. Chernenko, B. J. Hattink, and A. Labarta, Phys. Rev. B **60**, 7085 (1999).