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RAMAN SCATTERING IN V_3Si , V_3Ge , Nb_3Sb , AND Cr_3Si : CORRELATION OF E_g OPTICAL PHONON LINEWIDTH WITH MAGNETIC SUSCEPTIBILITY

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Abstract.— Raman scattering measurements of the E_g optical phonon in V_3Si and Nb_3Sn show it to have an anomalous width, temperature dependence, and asymmetric lineshape. We have observed a similar, although weaker, anomaly in V_3Ge . The E_g and T_{2g} phonons in Nb_3Sb and the T_{2g} phonon in Cr_3Si show no anomalous behavior and can be understood in terms of simple anharmonic interactions. In Cr_3Si the E_g phonon has an anomalous width, shape, and temperature dependence (similar to V_3Ge) in spite of its low electronic density of states and temperature independent magnetic susceptibility. A linear correlation is shown to exist between magnetic susceptibility and E_g mode linewidth, Γ , in V_3Si , Nb_3Sn , and V_3Ge . With the aid of a simple model, most of the features of the Raman data can be understood in terms of direct coupling of the E_g phonon to interband electronic transitions between the very flat bands emanating from the Γ_{12} level in these compounds. These results indicate that direct coupling of the E_g optical phonon to the Γ_{12} bands plays a major role in the splitting of the cubic Γ_{12} subband $N(E)$ peak by the dimerization of the transition metal sublattice.

1. **Experiment.**— A single crystal of V_3Ge ($T_c=6.3K$) was grown from buttons of stoichiometric arc-melted starting material by the Czochralski technique.^{1/} The single crystal of Cr_3Si ($T_c<0.015K$) was grown by zone refining pressed stoichiometric powder mixtures of Chromium and Silicon.^{2/} [100] surfaces were spark cut from these samples and then mechanically polished with alumina. For the investigation on Nb_3Sb , a large single crystal was grown by closed tube vapor transport with iodine as a transporting agent. The Raman measurements were performed on a high quality, smooth, as-grown [110] face of the crystal. Laser light of 514-nm wavelength was incident at a pseudo-Brewster angle of 70° , collected in a direction normal to the surface, and analyzed with a home-built double monochromator employing standard photon counting electronics. Cooling was provided by either flowing cold He gas in a modified "Heli-Tran" system or a liquid He Janis cryostat. True sample temperatures were determined from Anti-Stokes/Stokes ratios of the E_g phonon.

2. **Results and Discussion.**— The E_g symmetry Raman spectra of V_3Ge taken at 340K and 50K are shown in Fig. 1. The phonon frequency hardens from 278. to 287. cm^{-1} and the linewidth increases from 37.2 to 69.6 cm^{-1} (FWHM) upon cooling. The solid lines are fits to a spectral function resulting from a coupled-mode theory, wherein the asymmetric lineshape is due to a Breit-Wigner-Fano interference between the discrete phonon and an electronic continuum.^{3/}

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Fig. 1 : E_g symmetry Raman spectra in V_3Ge at 340K and 50K. The solid lines are fits to the data (see text).

are weakly damped and harden and narrow upon cooling. This is typical behavior for phonons whose self-energy is dominated by anharmonic phonon-phonon interactions. The E_g phonon in Cr_3Si is not so well understood. It appears to harden from 310. to 320. cm^{-1} and broaden from 40. to 60. cm^{-1} upon cooling from 300K to 70K. However, additional structure in the region of the phonon, possibly due to electronic Raman scattering, complicates the analysis.

The electronic nature of the E_g phonon damping in V_3Si , Nb_3Sn , and V_3Ge is further demonstrated by plotting their magnetic susceptibility, χ , versus their corresponding E_g mode linewidth at the same temperature, for several temperatures. This is shown in Fig. 2 for the temperature range 400K to 20K, where a strong correlation is seen to exist, with the V-based compounds following one relationship and Nb_3Sn a separate one. Such a division of V-based and Nb-based A-15 compounds into two families, with $\eta_{Nb_3Sn} > \eta_{V_3B}$, where $\eta = N(E_F) \langle I^2 \rangle$, $\langle I^2 \rangle$ being the Fermi surface averaged electron-phonon matrix element squared, has already been pointed out by Klein, et. al.^{8/} The ratio of the initial slopes of the two curves (indicated by the solid lines through the data in Fig. 2) is in good agreement with the ratio of $\langle I^2 \rangle_{Nb_3Sn} / \langle I^2 \rangle_{V_3Si}$ as calculated by Klein, et. al.^{8/} The temperature dependence of the magnetic susceptibility is commonly ascribed to thermal repopulation of electronic energy levels near a sharp peak in the electronic density of states. The strong correlation between the magnetic susceptibility and the E_g phonon linewidth, shown in Fig. 2, suggests that electron-phonon interactions (as opposed to anharmonic phonon-phonon interactions), subject to the same thermal repopulation effects as the magnetic susceptibility, are responsible for the temperature dependence of

We propose that the dominant damping mechanism for the E_g mode in this A-15 and in V_3Si ^{3,4/} and Nb_3Sn ^{4/} consists of inter-band processes wherein the E_g mode decays into electron-hole pairs in the Γ_{12} bands. These bands are within an energy comparable to the E_g phonon energy of the Fermi level throughout large regions of the Brillouin Zone.^{5/} Due to symmetry based selection rules, the T_{2g} phonon in Nb_3Sn cannot interact with these bands and it has indeed been observed to be relatively weakly damped.^{4/}

In Nb_3Sb ^{6/} and Cr_3Si ^{7/} the E_g phonon cannot interact with electrons in the Γ_{12} bands since they are ~ 0.35 eV below the Fermi level and hence completely filled. Accordingly, the E_g phonon in Nb_3Sb and the T_{2g} phonons in both Nb_3Sb and Cr_3Si

Fig. 2 : Correlation of χ vs. Γ for V_3Si , Nb_3Sn , and V_3Ge . Raman data for Nb_3Sn taken from ref. 4. data for V_3Si , V_3Ge , and Nb_3Sn taken from refs. 9, 9', and 10, respectively.

the E_g phonon linewidth and frequency in these compounds. Indeed, a simple model of the interaction and the Γ_{12} bands is able to account quantitatively for the linewidth temperature dependence. Our model will be discussed more fully in a future publication. These results are in accord with the recent conclusions of Mattheiss and Weber^{11/} that dimerization of the transition metal sublattice is the primary driving mechanism for the martensitic transition.

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