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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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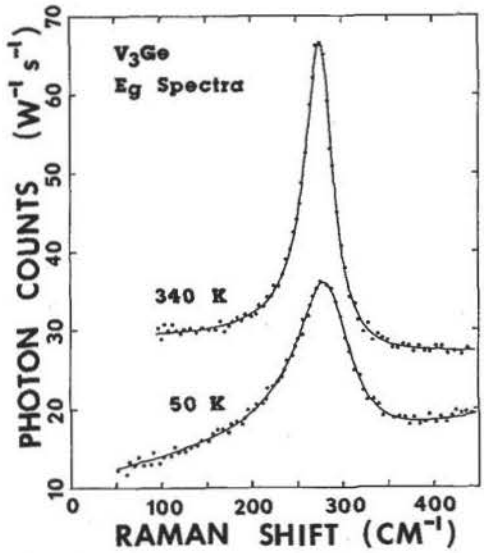

Fig. 1 : E_g symmetry Raman spectra in V_3Ge at 340K and 50K. The solid lines are fits to the data (see text).

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 interband 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 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, X, 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 seperate one. Such a division of V-based and Nb-based A-15 compounds into two families, with $\eta_{Nb_3B} > \eta_{V_3B}$, 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

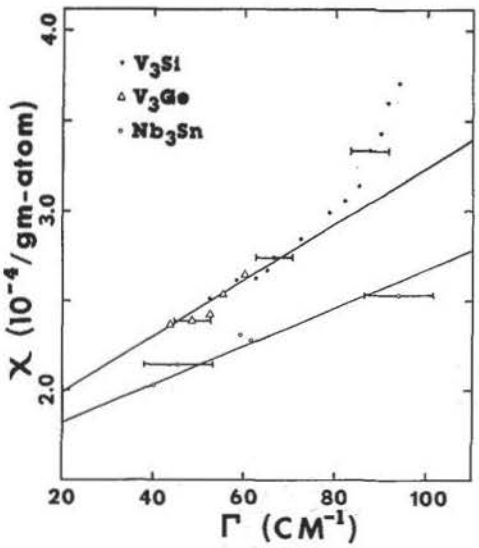

Fig. 2 : Correlation of vs. for V_3Si, Nb_3Sn, and V_3Ge. Raman data for Nb_3Sn taken from ref. 4. data for V_3Si, V_3Ge, and Nb_3Sn taken from refs. 9, 9, and 10, respectively.

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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