

## Brief Reports

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### “Clustering modes” in the vibrational spectra of $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ alloys

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The spectral densities of states for phonons in  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  alloys are predicted using the recursion method and are compared with Raman scattering data. The “clustering mode” observed by Amirtharaj *et al.* is shown to be an “alloy mode” associated with vibrations of a Te atom bonded to one Cd atom and three Hg atoms.  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  is shown to be a “two-mode” alloy vibrationally.

Recent Raman scattering data<sup>1</sup> for  $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$  substitutional crystalline alloys revealed a vibrational mode at  $135\text{ cm}^{-1}$  that is neither Hg-like nor Cd-like. This “clustering mode” was thought to be caused by either nonrandom clusters in the alloy or by vacancies or vacancy-complexes. Motivated by these data, we have executed calculations of the densities of vibrational states of  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  random substitutional alloys, using the recursion method.<sup>2</sup> As a result (i) we have determined that the clustering mode<sup>1</sup> is attributable to clusters of one Cd atom and three Hg atoms bonded to the same Te; there is no need to invoke vacancies or nonrandom clusters to describe the observed mode; and (ii) we have found that  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  is a “two-mode” alloy<sup>3</sup> in that the optical phonons of HgTe and CdTe persist for all  $x$ .

The calculations employ the recursion method,<sup>2</sup> using a model with only short-ranged forces.<sup>4-6</sup> The method and model are essentially the same as those used for  $\text{Al}_x\text{Ga}_{1-x}\text{As}$  alloys<sup>6</sup> and  $(\text{GaAs})_{1-x}\text{Ge}_{2x}$  alloys,<sup>7</sup> and so

TABLE I. Force-constant parameters (in units of dyn/cm) and masses (in units of  $10^{-24}\text{ g}$ ) for HgTe and CdTe, in the notation of Ref. 6.

	HgTe	CdTe
$\alpha$	-15 875	-19 806
$\beta$	-14 635	-18 469
$\lambda_c$	5302	5304
$\lambda_a$	2579	1360
$\mu_c$	-2486	-5281
$\mu_a$	-6650	-2704
$\nu_c$	-4830	-6954
$\nu_a$	-2930	2105
$m_c$	333.1	186.6
$m_a$	211.9	211.9

we do not repeat the formalism here. The recursion method is a Green’s-function theory, and, as such, has elements in common with the work of Talwar and Vandevyver.<sup>8</sup> The force constants used for HgTe and CdTe were fit to neutron-scattering data,<sup>9,10</sup> and are given in Table I. They produce good phonon-dispersion curves

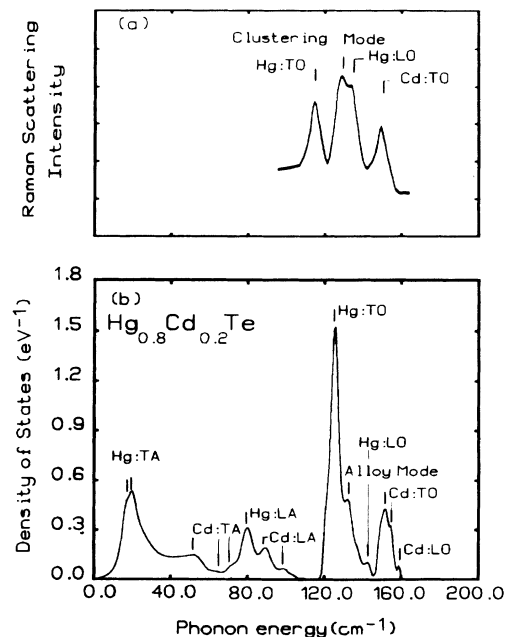


FIG. 1. (a) The Raman scattering data of  $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ , according to Ref. 1, in comparison with (b) the calculated density of states of  $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ . The notation Cd:TA, for example, indicates a transverse optic phonon of CdTe. The data were taken for the  $\langle 1\bar{1}0 \rangle$  face at 77 K and are the curve 2a of Fig. 2 from Ref. 1. Principal spectral features are identified on the figure.

and vibrational densities of states.<sup>11,12</sup>

Here we report the recursion calculations for  $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ , for which Raman scattering data are available.<sup>1</sup> Principal peaks of the spectra of HgTe and CdTe are identified in Fig. 1(a), using standard notation. Note, in particular, the clustering mode, whose origin has been a subject of speculation.<sup>1</sup> In addition, an "alloy mode," which appears in the spectra of neither HgTe nor CdTe, is indicated in Fig. 1(b). Such alloy modes are associated, in general, with vibrations of specific clusters of atoms in  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ . The identification of such peaks in the spectra with particular clusters of atoms is obtained by choosing various specific miniclusters,<sup>6,7</sup> and by computing the local densities of states for one atom at the center of such a minicluster.

Comparing our theoretical results with the Raman scattering data of Amirtharaj *et al.*<sup>1</sup> (Fig. 1), we find good agreement with the Cd-like optic mode at  $156\text{ cm}^{-1}$  and the two Hg-like modes at  $122$  and  $144\text{ cm}^{-1}$ , respectively. The experimental clustering mode is simply an alloy mode<sup>6</sup> due to Te atoms bonded to three Hg atoms and one Cd atom. There is no need to invoke nonrandomness in the alloy or to postulate the existence of vacancies in high concentrations.

We find that the optic modes of HgTe and CdTe persist for all alloy compositions  $x$  in  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ . This is to be expected on the basis of the criterion of Onodera

and Toyozawa:<sup>13</sup> because the masses of Hg and Cd are so different, the spectrum of  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  exhibits two-mode persistent behavior, and the optical phonon bands of HgTe and CdTe persist for all compositions  $x$  in the alloy  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ . Note that the model on which the recursion calculations are based contains no long-ranged forces; hence the longitudinal- and transverse-optic phonons are degenerate in this model at  $\mathbf{k}=\mathbf{0}$ , the Brillouin zone center. Since the experimental longitudinal-transverse optic phonon splitting at  $\mathbf{k}=\mathbf{0}$  is small, the omission of the long-ranged Coulomb forces responsible for the splitting does not alter the essential physics or impede the identification of the principal spectral features.

In summary, recursion-method calculations of the densities of states of  $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$  alloys provide a natural identification of the clustering mode with vibrations of a complex involving three Hg atoms and one Cd atom bonded to the same Te atom. A complete discussion of the formalism and the phonon spectra of these and other interesting alloys will be published soon.<sup>11</sup>

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<sup>2</sup>R. Haydock, *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1980), Vol. 35, p. 215.

<sup>3</sup>G. Lucovsky, K. Y. Cheng, and G. L. Pearson, *Phys. Rev. B* **12**, 4135 (1975).

<sup>4</sup>The model is the same as that of Ref. 5, except that the long-ranged forces are omitted, as in Ref. 6.

<sup>5</sup>R. Banerjee and Y. P. Varshni, *Can. J. Phys.* **47**, 451 (1969).

<sup>6</sup>A. Kobayashi, J. D. Dow, and E. P. O'Reilly, *Superlatt. Microstruct.* **1**, 471 (1985).

<sup>7</sup>A. Kobayashi, K. E. Newman, and J. D. Dow, *Phys. Rev. B* **32**, 5312 (1985).

<sup>8</sup>D. N. Talwar and M. Vandevyver, *J. Appl. Phys.* **56**, 1601 (1984).

<sup>9</sup>H. Kepa, W. Gebicki, T. Giebultowicz, B. Buras, and K. Clausen, *Solid State Commun.* **34**, 211 (1980).

<sup>10</sup>J. M. Rowe, R. M. Nicklow, D. L. Price, and K. Zanio, *Phys. Rev. B* **10**, 671 (1974).

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<sup>12</sup>The vibrational density of states is defined as follows:  $\sum_{s,b,i} |\langle n,b,i | s \rangle|^2 \delta(\omega - \omega_s)$ , where  $n$  labels the central unit cell of a 1000-atom cluster,  $b$  denotes either  $a$  (anion) or  $c$  (cation) in that cell,  $i$  runs over  $x$ ,  $y$ , and  $z$ , and  $s$  labels the normal modes of the cluster:  $\omega_s$  are the eigenfrequencies and  $|s\rangle$  are the eigenvectors of the cluster. We used 51 levels of recursion, setting the fifty-first continued-fraction coefficient to zero.

<sup>13</sup>Y. Onodera and Y. Toyozawa, *J. Phys. Soc. Jpn.* **24**, 341 (1968).