

range by a curve with characteristic width exponent  $\mu = 0.63 \pm 0.03$ .

Of equal importance, with explanation of the reflectance experiments, are the purely theoretical questions concerning the fundamental role of the external gravitational field. Although detailed analysis must await our fuller exposition of the general interfacial theory, we remark that Eqs. (5) and (6) predict that the diffuse planar interface diverges in width logarithmically as  $g \rightarrow 0$  ( $T$  fixed), and that  $\gamma$  suffers simultaneously a bounded anomaly, also of logarithmic character.

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\*A portion of the underlying theory was presented by one of us (F.P.B) in the Proceedings of the Thirty-Eighth National Colloid Symposium, 11-13 June 1964, Austin, Texas (to be published).

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<sup>1</sup>G. H. Gilmer, W. Gilmore, J. Huang, and W. W. Webb, Phys. Rev. Letters **14**, 491 (1965).

<sup>2</sup>J. D. van der Waals, Z. Physik. Chem. (Leipzig) **13**, 657 (1894). The extension of the theory to binary sys-

tems is due to J. W. Cahn and J. E. Hilliard, J. Chem. Phys. **28**, 258 (1958).

<sup>3</sup>This notion in the microscopic regime seems first to have appeared in M. V. Smoluchowski, Ann. Physik **25**, 205 (1908).

<sup>4</sup>C. Eckart, Phys. Rev. **35**, 1303 (1930).

<sup>5</sup>For profile (1), distance  $L$  is related to the corresponding second moment by  $\langle z^2 \rangle^{1/2} = (3^{1/2}\pi/12)L = 0.453L$ . Although the van der Waals density-gradient development gives  $\mu = \frac{1}{2}$ , the theory leading to (1) may be cast into a form freeing it from the requirements of "classical" bulk phase behavior. In terms of critical density  $\rho_c$ , isothermal compressibility  $\kappa_T$ , and surface tension  $\gamma$ , one finds  $L = 6[\rho_c/(\rho_1 - \rho_2)]^2 \kappa_T \gamma$ , so presently available experimental results on  $\gamma$  and  $\kappa_T$  yield  $\mu$  in the more acceptable range 0.6-0.7. For notational economy, the single-component liquid-vapor case is assumed throughout.

<sup>6</sup>R. H. Fowler, Proc. Roy. Soc. (London) **159**, 229 (1937).

<sup>7</sup>D. Atack and O. K. Rice, Discussions Faraday Soc. **15**, 210 (1953). Upon making requisite gravitational-field corrections,  $\gamma \propto (T_c - T)^{1.29}$  for this mixture. This temperature dependence is consistent with the prediction of our "nonclassical" density-gradient-type theory  $\gamma \propto (\rho_1 - \rho_2)^2 / \kappa_T^{1/2}$ .

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## OBSERVATION OF RESONANT LATTICE MODES BY INELASTIC NEUTRON SCATTERING\*

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This Letter reports the observation by inelastic neutron scattering of resonant lattice modes due to a small concentration of W atoms in a Cr host crystal.

The modification of the modes of vibration of a lattice due to the substitution of heavy or light impurities for some of the host atoms has been the subject of much recent discussion. The scattering of neutrons by such a defect structure has been considered by Krivoglaz,<sup>1</sup> Kagan and Iosilevskii,<sup>2</sup> and Brout and Visscher.<sup>3</sup> More recently, Elliott and Maradudin<sup>4</sup> have given a detailed discussion of neutron scattering by the defect modes and have shown, with explicit calculations, how information about such modes can be obtained from the experiments. It was this work which provided the stimulus for our experiments.

The type of lattice mode which occurs near a defect depends on whether the mass of the impurity  $M'$  is less or greater than  $M$ , that of the host atoms. In the former case, a localized mode appears with a frequency above the frequency distribution of the host lattice, while in the latter there is a resonant mode within the allowed frequency range of the host. This mode is characterized by a large amplitude of vibration at the defect, decaying into a normal lattice mode of the host at large distances. The frequencies and lifetimes of phonons with frequencies near that of the resonant mode are considerably affected by the presence of the defects. Elliott and Maradudin<sup>3</sup> show that, for a cubic lattice, a small concentration  $c$  of impurities which do not affect the interatomic force constants causes a frequency shift  $\Delta$  and

broadening  $\gamma$  of the one-phonon neutron groups, given by

$$\Delta(q) = c V_1[\omega(q)]/2\omega(q), \quad \gamma(q) = c V_2[\omega(q)]/\omega(q), \quad (1)$$

where

$$V_1(\omega) + iV_2(\omega) = \epsilon \omega^2 \left\{ \left[ 1 - \epsilon \omega^2 \int \frac{g(\omega') d\omega'}{\omega^2 - \omega'^2} \right] + \frac{1}{2} i \pi \epsilon \omega g(\omega) \right\} \times \left\{ \left[ 1 - \epsilon \omega^2 \int \frac{g(\omega') d\omega'}{(\omega^2 - \omega'^2)^2} \right]^2 + \frac{1}{4} \pi^2 \epsilon^2 \omega^2 g^2(\omega) \right\}^{-1}. \quad (2)$$

In this equation  $\epsilon = (M - M')/M$ ,  $g(\omega)$  is the density of states in the unperturbed lattice, and the principal part of the integrals is to be taken.

The width reaches a maximum at the resonant frequency, for which  $V_1 = 0$ , and for a Debye model, this maximum width is given approximately by<sup>3</sup>

$$\gamma/\omega = (2c/3\pi)[3(M'/M - 1)]^{3/2}. \quad (3)$$

At low frequencies, the frequency shift is negative and is determined by the change in the mean mass of the crystal:

$$\Delta(q) = \frac{1}{2} c \epsilon \omega(q). \quad (4)$$

At the resonant frequency the shift changes sign and the relative total change across the resonance is of the order of  $\frac{1}{2} c \epsilon$ . At high frequencies the shift again becomes slightly negative.

A dilute alloy of W in Cr is well suited for studying these phenomena. Measurements of the phonon spectra in the pure metals<sup>5,6</sup> have revealed very similar force-constant distributions, thus satisfying the primary assumption of the theory. W is a very heavy atom, so that  $\epsilon$  for this system is  $-2.54$  and the resonant modes should be well developed. We have therefore studied the lower transverse [110] phonon branch in single crystals of Cr and a Cr-3.0-at.% W alloy by inelastic neutron scattering, using the constant- $\bar{q}$  method. These experiments were carried out at the DR3 reactor of the Danish Atomic Energy Commission Research Establishment. The observed neutron groups are shown in Fig. 1, while the energy shifts are plotted as a function of the phonon energies in pure Cr in Fig. 2. There is clear evidence of a resonant mode near 21 meV. There is a large change in the frequency shift, of the right order of magnitude, while the neutron groups broaden perceptibly near  $q = 0.694$ . Equation (3) predicts a maximum width of about 3

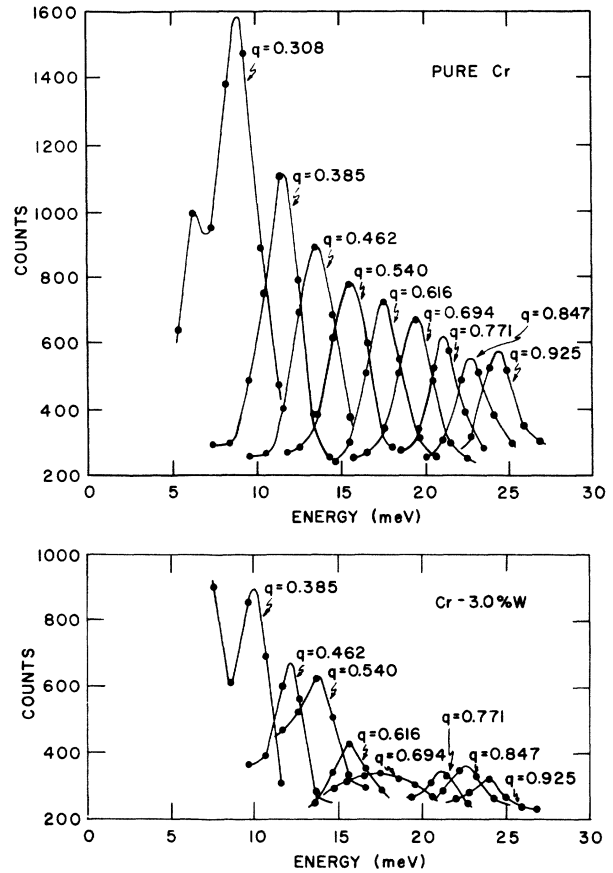


FIG. 1. Neutron groups in pure Cr and the Cr-W alloy at room temperature. The phonon wave vector  $\bar{q}$  is in the [110] direction and is measured in reciprocal Å.

meV, which is roughly the magnitude observed. There are, however, certain discrepancies with the theory. The positive frequency shift appears to be very small, while the maximum broadening occurs where  $\Delta$  is large and negative, rather than where it is zero. Similar effects and more pronounced discrepancies with the theory have recently been observed in a Cu-9.3-at.% Au alloy by Svensson, Brockhouse, and Rowe.<sup>7</sup> These discrepancies may be ascribed

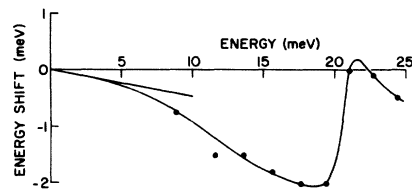


FIG. 2. Energy shifts in the Cr-W alloy as a function of the phonon energy in pure Cr. The straight line through the origin is a plot of Eq. (4).

to the relatively large concentration of impurities and changes in the force constants near them.

The resonant mode occurs at approximately one-half of the maximum phonon energy of 42 meV in Cr. From Eq. (2) and the phonon dispersion curves<sup>5</sup> it should be possible to calculate the frequency shifts to compare with experiment. We have observed evidence for resonant modes at approximately the same frequency in other branches of the phonon spectrum, and we are extending our preliminary experiments to make a detailed study of these.

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<sup>1</sup>M. A. Krivoglaz, Zh. Eksperim. i Teor. Fiz. **40**, 567 (1961) [translation: Soviet Phys.—JETP **13**, 397 (1961)].

<sup>2</sup>Yu. Kagan and Ya. Iosilevskii, Zh. Eksperim. i Teor. Fiz. **44**, 1375 (1963) [translation: Soviet Phys.—JETP **17**, 925 (1963)].

<sup>3</sup>R. Brout and W. M. Visscher, Phys. Rev. Letters **9**, 54 (1962).

<sup>4</sup>R. J. Elliott and A. A. Maradudin, In Inelastic Scattering of Neutrons (International Atomic Energy Agency, Vienna, 1965).

<sup>5</sup>S. H. Chen and B. N. Brockhouse, Solid State Commun. **2**, 73 (1964).

<sup>6</sup>H. Bjerrum Møller and A. R. Mackintosh, in Inelastic Scattering of Neutrons (International Atomic Energy Agency, Vienna, 1965).

<sup>7</sup>E. C. Svensson, B. N. Brockhouse, and J. M. Rowe, to be published.

## OPTICAL ABSORPTION BY EXCITONS IN A STRONG ELECTRIC FIELD

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In this Letter we describe a nonperturbative treatment of a hydrogenic two-body system in the presence of a strong electric field. Our major results are (1) the demonstration that asymptotically for large negative energies the presence of the Coulomb interaction does not materially alter the optical absorption in an electric field alone, and (2) the calculation of the widths and energies of the low-energy exciton peaks as a function of the electric field strength. A number of speculations concerning the above results have appeared in the literature.<sup>1,2</sup> We present their first quantitative calculation. Such a calculation is needed to distinguish structure due to excitons from that due to critical points in electric-field-modulated optical reflectance<sup>2,3</sup> and transmission<sup>4</sup> data. In the course of our analysis, we perform the first exact solution of Schrödinger's equation for a potential, other than a square barrier, which exhibits bound states in the continuum.

If the interaction between the optical field and the solid is treated in first-order perturbation theory, then in the absence of other external fields it has been shown by Elliot<sup>5</sup> that the calculation of the optical-absorption tran-

sition probability reduces to evaluating the matrix element

$$M = \sum_{\vec{k}_e, \vec{k}_h} \Psi_{\vec{k}_e, \vec{k}_h, j, j'}^{\vec{K}, n} I_{jj'}(\vec{k}_e, \vec{k}_h, \vec{\xi}, \vec{q}), \quad (1a)$$

$$I_{jj'}(\vec{k}_e, \vec{k}_h, \vec{\xi}, \vec{q}) = \int d^3r \psi_{\vec{k}_e, j}^+(\vec{r}) e^{i\vec{q} \cdot \vec{r}} \left( \frac{i e \hbar}{m} \right)_{\vec{\xi}} \cdot \vec{\nabla}_{\vec{k}_h, j'}(\vec{r}), \quad (1b)$$

in which  $\vec{q}$  is the wave vector and  $\vec{\xi}$  the polarization vector of the light wave;  $\vec{k}_e$  and  $\vec{k}_h$  are the wave vectors of the electron and hole from bands  $j$  and  $j'$ , respectively; the  $\psi_{\vec{k}, j}$  are their one-electron wave functions, and  $\Psi$  is the Fourier transform of an exciton wave function. The imposition of an external electric field alters both the expansion coefficients,  $\Psi$ , and the one-electron wave functions,  $\psi$ . The effects of the electric field on  $\psi_{\vec{k}, j}$ , as reflected in the integral (1b), are thought to be small.<sup>6</sup> As the integral is customarily treated as a variable parameter used to fit the experimental data,<sup>7</sup> we do not discuss it further.

For direct, allowed transitions between two