

although believable, is not rigorous. It is somewhat unfortunate that in Refs. 3 and 4 the argument is never given nor is any statement made of how rigorously the result is considered to have been derived.

⁶T. Kato, *Commun. Pure Appl. Math.* **10**, 151 (1957). In the zeroth-order problem (the one associated with W_0) the potential has a cusp, so that the correct solution should be continuous with a continuous derivative. Kato's proofs do not rigorously apply to the eigenfunctions of the continuous spectrum, but as Bingel has observed in the one-body Coulomb (i.e., hydrogenic) problem, the cusp condition applies to the eigenfunctions of the continuous as well as those of the discrete spectrum. Cf. W. A. Bingel, *Z. Naturforsch.* **18a**, 1249 (1963).

⁷R. K. Peterkop, *Zh. Eksperim. i Teor. Fiz.* **43**, 616 (1962) [translation: *Soviet Phys.-JETP* **16**, 442 (1963)].

⁸Physically it would appear preferable to use an outgoing wave for the inner particle in place of the regular solution. However, arguments of stationary phase indicate that the ingoing parts of the regular function

cancel themselves out asymptotically. Although we shall question the stationary-phase results below, it is unlikely that this aspect of what it implies is incorrect. I am indebted to R. K. Peterkop for an informative letter.

⁹A. Temkin, *Phys. Rev.* **130**, 126 (1962).

¹⁰Strictly speaking, what we have shown is that if $C_n \propto n^{-\gamma}$, then $\gamma = \frac{3}{2}$. Furthermore, we require the extrapolation of $n^{-3/2}$ to $k^{3/2}$ for the ionization coefficients above threshold, which has not been proved. The demonstration that the ionization coefficients having the dependence $k^{3/2}$ leads to an $E^{3/2}$ law is taken from K. Omidvar, private communication. A more detailed derivation of the zeroth-order threshold dependence will be given in a future report.

¹¹R. K. Peterkop, private communication.

¹²J. W. McGowan, M. A. Fineman, E. M. Clarke, and H. P. Hanson, to be published. Wannier, Ref. 3, also derived a nonlinear threshold dependence. The experimental nonlinearity that is in question here occurs within a tenth of an electron volt of threshold.

COLLECTIVE MOTION IN LIQUID ARGON†

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Recently Chen *et al.*¹ have observed well-defined inelastic peaks in the energy distribution of slow neutrons scattered by liquid argon. These inelastic peaks were not resolved by earlier experiments.^{2,3} In this note we present independent evidence that the observed inelastic peaks are a real manifestation of collective motion in liquid argon.

Rahman⁴ has carried out a classical molecular-dynamics calculation of the Van Hove correlation functions $G_S(r, t)$ and $G_d(r, t)$ for liquid argon. We have used his results to compute the expected inelastic neutron scattering under the conditions of Ref. 1 and find a structure in the energy distribution of scattered neutrons similar to that reported by Chen *et al.*

We need the double Fourier transforms $S_S(\kappa, \omega)$ and $S_d(\kappa, \omega)$ of the Van Hove correlation functions. This is not feasible on a completely numerical basis from Rahman's results,

but Rahman has already given us enough information to carry out the spatial transform analytically. He has found that his data are well fitted by a delayed convolution approximation. For the intermediate scattering function [transform of $G(r, t)$ with respect to r] this implies

$$F_d(\kappa, t) = [S(\kappa) - 1] F_S(\kappa, t'), \quad (1)$$

where $S(\kappa)$ is the usual structure factor. The time t' is given by Rahman as

$$t' = t - \tau [1 - \exp(-t/\tau) - (t^2/\tau^2) \exp(-t^2/\tau^2)] \quad (2)$$

with $\tau = 1.0 \times 10^{-12}$ sec. It should be emphasized that this is a fit to the computed correlation functions, and not an approximate theoretical construct. The physical origin of this approximation is not well understood. Furthermore, $F_S(\kappa, t)$ is almost a Gaussian function of κ and can be written in terms of the spatial moments of $G_S(r, t)$ in the form

$$F_S(\kappa, t) = \exp[-\kappa^2 \gamma_1(t)] \left\{ 1 + \alpha_2(t) \frac{[\kappa^2 \gamma_1(t)]^2}{2!} - [\alpha_3(t) - 3\alpha_2(t)] \frac{[\kappa^2 \gamma_1(t)]^3}{3!} + [\alpha_4(t) - 4\alpha_3(t) + 6\alpha_2(t)] \frac{[\kappa^2 \gamma_1(t)]^4}{4!} - \dots \right\}, \quad (3)$$

where $\gamma_1(t)$ is one-sixth the mean-square displacement and $\alpha_n(t) = [(\langle r^{2n} \rangle / c_n \langle r^2 \rangle^n) - 1]$ with $c_n = 1 \times 3 \times 5 \times \dots \times (2n + 1) / 3^n$. Equation 3 is due to Nijboer and Rahman⁵ who have used it to analyze the incoherent inelastic neutron scattering from liquid argon by numerical Fourier transformation. Their results show no structure in the energy spectrum of scattered neutrons, as would be expected from the gaslike behavior of the calculated mean-square displacement.

That the situation might be different for coherent scattering is indicated by examining $\gamma_1(t')$ as a function of t in Fig. 1. A mean-square displacement of this form is well known to give some structure to incoherent inelastic scattering. The quantity that we finally compute is

$$\frac{d^2\sigma}{d\Omega d\lambda} = \frac{4\pi\hbar\lambda_i}{m\lambda^4} \exp\left[-\left(\frac{\hbar^2\kappa^2}{8mkT} + \frac{\hbar\omega}{2kT}\right)\right] \times \int_0^\infty dt \cos\omega t [\langle a^2 \rangle F_s(\kappa, t) + \langle a \rangle^2 F_d(\kappa, t)], \quad (4)$$

where λ_i is the incident neutron wavelength, $\hbar\omega$ is the energy gain by a neutron, and $\langle a \rangle^2$, $\langle a^2 \rangle$ are the usual scattering lengths for liquid argon. We have chosen $[\langle a \rangle^2 / \langle a^2 \rangle] = 0.675$. In Eq. (4), we have also included a quantum mechanical correction factor.⁶ This factor varies (for $\theta = 60^\circ$) from 0.55 to 0.89 over the inelastic peak and from 0.28 to 1.03 as the final wavelength, λ , is varied from 2.2 to 8.0 Å. This factor does not affect the qualitative features of our results, but its size indicates that a more complete treatment of quantum mechanical corrections is needed.

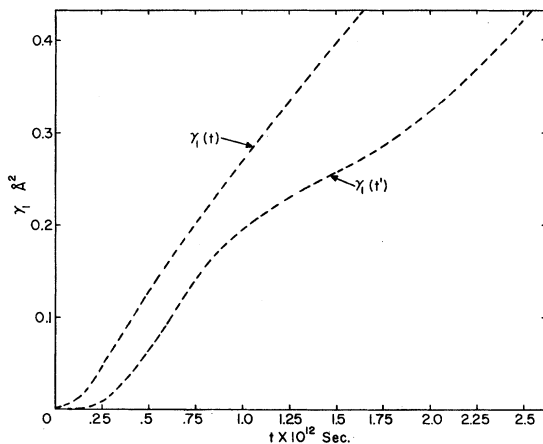


FIG. 1. $\gamma_1(t)$ and $\gamma_1(t')$ as functions of t .

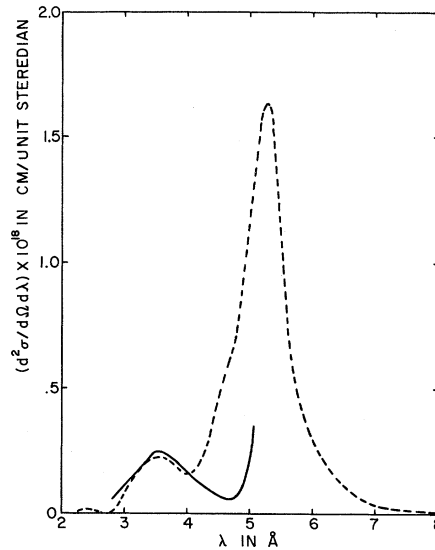


FIG. 2. Comparison of experiment of Chen *et al.* (solid curve) with the present calculation (dashed curve) at $\theta = 60^\circ$.

In Fig. 2, we show the wavelength distribution of scattered neutrons for 5.3-Å incident neutrons scattered through an angle of 60° . We also show the results of Chen *et al.*¹ The qualitative agreement is seen to be good. In Fig. 3, we show the same comparison at a scattering angle of 75° . We note that the experimental peak has become better defined

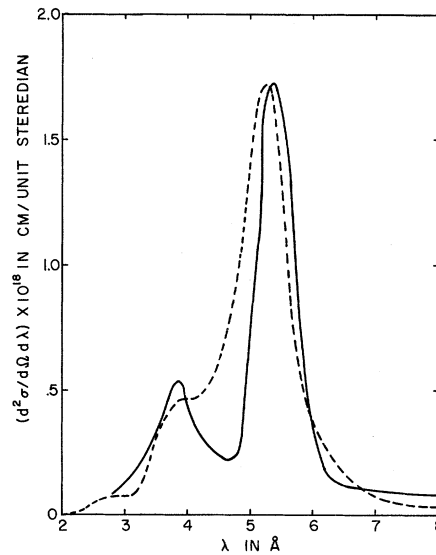


FIG. 3. Comparison of experiment of Chen *et al.* (solid curve) with the present calculation (dashed curve) at $\theta = 75^\circ$. Values of elastic maximum in two cases are matched.

whereas the computed peak is more smeared out. A possible source of the discrepancy is that the experiments were done at a temperature of 85°K whereas Rahman's computer experiments were at 94.4°K. We have used Henshaw's neutron-diffraction measurements⁷ at 84°K for $[\langle a \rangle^2 / \langle a^2 \rangle]$ and $S(\kappa)$, but our results are not particularly sensitive to this quantity since the momentum transfer κ is not a rapidly varying function of the final neutron wavelength.

We have not chosen to interpret the inelastic peaks in terms of a dispersion relation for the cooperative modes since the location of the peak in κ and ω is quite sensitive to the way in which the data are plotted. In fact, if we plot $S(\kappa, \omega)$ as a function of ω for fixed κ , we get only a rather flat shoulder on the curve rather than a well-defined peak. Even this structure is not present, however, in $S_S(\kappa, \omega)$. Thus the molecular-dynamics calculations of Rahman support the neutron-scattering experiments in predicting an observable effect of collective motion in liquid argon. We still do not have any theoretical framework which al-

lows this motion to be quantitatively described in terms of any well-defined elementary excitations.

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EFFECT OF UNIAXIAL STRESS ON MAGNON SIDEBANDS IN MnF₂

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Recently, a sideband of an optical transition in the absorption spectrum of MnF₂ was identified as arising from the simultaneous creation of an exciton and a magnon.¹ In this paper we report wave functions for the excitons² and selection rules for the coupling of the excitons to the magnons, as derived from optical absorption measurements of crystals of MnF₂ stressed uniaxially along the 001 and 110 directions. The zero-stress spectrum of the transition ${}^6A_{1g} \rightarrow {}^4T_{1g}$ in MnF₂, as reported in Ref. 1, consists of two sharp weak magnetic dipole lines $E1$ (at 18 419.6 cm⁻¹) and $E2$ (at 18 436.6 cm⁻¹), and three broad, relatively intense electric dipole lines $\sigma 1$ (at 18 477.1 cm⁻²), $\sigma 2$ (at 18 485.3 cm⁻¹), and $\pi 1$ (at 18 460.8 cm⁻¹). The magnetic dipole lines are observed in σ polarization only, and were attributed in Ref. 1 to exciton transitions; the polarizations of the electric dipole lines are

indicated in their identifying symbols. One of these lines, $\sigma 1$, was identified in Ref. 1 as a magnon sideband of $E1$ by the similarity of the shape and temperature dependence of the peak frequency to previous measurements of the magnon spectrum by other techniques.

Our measurements show that [001] stress shifts $E1$, $\pi 1$, and $\sigma 1$ by the same amount, while shifting $E2$ and $\sigma 2$ by a much smaller amount. This result confirms the identification in Ref. 1 of $\sigma 1$ as a sideband of $E1$, and, in addition, indicates $\pi 1$ to be also a sideband of $E1$, and $\sigma 2$ a sideband of $E2$. These results are summarized on the left-hand side of Fig. 1.

Whereas [001] stress merely shifts the absorption lines without splitting them, [110] stress splits each line into two. This result is to be expected since [001] stress affects the two Mn sublattices in the same way, while [110] stress acts along an x direction³ of one sublattice