Mode-coupling effects in dense hydrogen gas

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A thermal-neutron scattering experiment on dense hydrogen gas at 120 K and 730 bar reveals that the measured incoherent dynamic structure factor is consistent with the predictions derived from the mode-coupling theory up to wave numbers of about 1 Å^{-1}.

I. INTRODUCTION

A few years ago Chen, Postol, and Sköld[1] performed neutron scattering experiments on hydrogen at temperatures $T=78$ and 293 K and pressures p ranging from 100 up to 2000 bar. Thus they determined the incoherent dynamic structure factor $S_s(k,\omega)$ as a function of frequency ω for wave numbers $0.35\ \text{Å}^{-1} \leq k \leq 0.8\ \text{Å}^{-1}$. Their aim was to find experimental evidence for the anomalous collective effects in fluids which are predicted by the mode-coupling theory.[2–4] They found that for sufficiently small k the half-width of $S_s(k,\omega)$ is roughly proportional to k^2 and, invoking Fick's diffusion law, identified the proportionality constant as the coefficient of self-diffusion, D. As a function of density, the measured values of D showed a behavior similar to that observed in systems of hard spheres.[5,6] Since for hard spheres the collective long-time tail effects in the velocity-autocorrelation function are necessary to describe the behavior of D properly,[6] Chen *et al.* thus found indirect evidence for the existence of mode-coupling effects in real fluids. In the present paper we investigate to what extent the predictions of the mode-coupling theory for $S_s(k,\omega)$ *itself*[7–9] can be observed *directly* in experimentally obtained neutron spectra for hydrogen. Thereto we have chosen a thermodynamic state for which we expect mode-coupling effects to be large and kinetic effects[1] to be small, while D is large enough to yield neutron spectra which are significantly broader than the actual widths of experimental resolution functions. We describe the experiment in Sec. II, the theory in Sec. III, and the results in Sec. IV. We discuss our results in Sec. V.

II. EXPERIMENT

We used the RKS-2 rotating-crystal time-of-flight spectrometer at the Delft 2-MW light-water swimming-pool reactor. The neutrons in the incident beam had a wavelength $\lambda_0=2.013$ Å corresponding to an energy of 20.19 meV. The cross section of the beam was $10\times2.5\ \text{cm}^2$. The sample consisted of 99.9% pure H_2 at 120 K and 730 bar, corresponding to a number density $n=0.020\,65\ \text{Å}^{-3}$.[10] The container[11] consisted of 30 parallel high-strength aluminum-alloy tubes, 1.3 mm apart, with 0.75 mm inner diameter and 0.25 mm wall thickness. The normal to the plane through the tubes made an angle of $+30°$ with the incident beam and of 0° with the plane through the beam and the detectors. Consequently 22 tubes, containing 0.94 cm^3 of H_2, were illuminated by the incident neutron beam. In part, this container and such a configuration were chosen to reduce the amount of container scattering to a minimum. The intensity on the sample was 2.10^4 neutrons/s and the measurement lasted for 110 h. The intensities $I(\varphi,\lambda)$ of scattered neutrons with wavelength λ were measured at 15 scattering angles φ from $-17.04°$ up to $+24.46°$, the smallest $|\varphi|$ being 7.72°. Thus, seen from any of the detectors the hydrogen in one particular container tube is never obscured by that in any other tube. As a consequence the corrections due to self-shielding in the sample are minimal for the measured neutron spectra. Using the methods of Ref. 12, the neutron spectra $I(\varphi,\lambda)$ were corrected for background radiation, container scattering, multiple scattering, self-shielding, and duty-cycle overlap and were normalized absolutely by means of a separate measurement on solid benzene at 120 K, for which the scattering properties are known, using the same arrangement and container. At each φ we found that of the total measured intensity, at most 3% of the neutrons were scattered multiply and that self-shielding corrections were smaller than 5%. Furthermore the scattering due to the container varied from 15% at large angles to 25% at small angles. We relate the corrected spectra $I_c(\varphi,\lambda)$ to $S_s(k,\omega)$ making the following approximations. First we neglect the contributions to $I_c(\varphi,\lambda)$ of coherent scattering and of rotational transitions in the H_2 molecules. Also, for the incoherent scattering contribution, the arguments of the spherical Bessel functions which appear in the relation between the differential cross section and $S_s(k,\omega)$ [cf. Eq. (11) of Ref. 1] are replaced by zero. A study on the basis of Refs. 13–15 reveals that the total error due to these approximations is smaller than 5% of $I_c(\varphi,\lambda)$ and is therefore acceptable compared to the estimated overall accuracy of about 5–10% obtained in the present experiment. Then[12,16,17]

$$I_c(\varphi,\lambda)=\frac{\pi\hbar\sigma_{\text{inc}}\lambda_0}{m}\int d\lambda'(\lambda')^{-4}S_s(k(\varphi,\lambda'),\omega(\lambda'))\times Q(k(\varphi,\lambda'),\omega(\lambda'))\boldsymbol{R}(\varphi,\lambda-\lambda')\ . \tag{1}$$

Here $\hbar$ is Planck's constant divided by 2π, σ_{inc} the effective cross section of a hydrogen molecule for incoherent

neutron scattering at 120 K, m the mass of a neutron

$$k(\varphi,\lambda)=2\pi\lambda_0^{-1}[1+(\lambda_0/\lambda)^2-2(\lambda_0/\lambda)\cos\varphi]^{1/2}\ ,$$

$$\omega(\lambda)=2\pi^2\hbar/(m\lambda_0^2)[1-(\lambda_0/\lambda)^2]\ ,$$

the quantum correction $Q(k,\omega)=\exp[\beta\hbar\omega/2-\beta\hbar^2k^2/(8M)]$ with M the mass of a hydrogen molecule and $\beta=1/(k_BT)$ with k_B Boltzmann's constant, while the normalized resolution function $R(\varphi,\lambda'')$ of the detector at angle φ [i.e., $\int d\lambda''\,R(\varphi,\lambda'')=1$] was obtained from the separate benzene measurement. In complete equilibrium at 120 K the sample consists of 67% ortho and 33% para hydrogen and therefore[13–15] $\sigma_{\rm inc}=147$ barn. In order to determine whether our sample had reached the equilibrium state, we proceeded as follows. During the full course of the experiment we measured the transmission of the sample for neutrons, which quantity depends on the actual ortho-para ratio in the fluid. In a separate experiment we measured the transmission also at 15 K. At 15 K we found changes in the transmission of about 10% per hour, meaning that the equilibrium state is reached in the course of a few hours. Since at 120 K the equilibrium state is reached *faster* than at 15 K and since we found no changes in the transmission at 120 K, our sample was in equilibrium. We finally remark that for $\lambda\simeq\lambda_0$ and for sharp resolutions [i.e., $R(\varphi,\lambda'')\simeq\delta(\lambda'')$] one has that $k\sim|\sin\frac{1}{2}\varphi|$, $\omega\sim\lambda$, and $I_c\sim S_s$ [cf. Eq. (1)].

III. THEORY

The mode-coupling theory predicts that, for small k, $S_s(k,\omega)$ is given by[9]

$$S_s(k,\omega)=\frac{1}{\pi}\frac{Dk^2}{\omega^2+(Dk^2)^2}+\frac{1}{\pi}\frac{1}{Dkk^*}\mathrm{Re}G((i\omega+Dk^2)/\delta Dk^2)+O(k^{-1/2}f(\omega/k^2))\ , \tag{2}$$

where the first term is due to Fick's diffusion law and is of order k^{-2} if ω is measured in units k^2 and the second term is the leading mode-coupling correction to this result and is of order $k^{-1}f(\omega/k^2)$. In Eq. (2), the wave number $k^*=16\pi\beta MnD^2$ and $\delta=D/(D+\nu)$ with $\nu=\eta/(Mn)$ the kinematic viscosity and η the shear viscosity. The complex function $G(z)$ is given by

$$G(z)=\tan^{-1}(z-1)^{-1/2}-(z-2)(z-1)^{1/2}z^2\ . \tag{3}$$

We remark that the Fourier transform $\hat\rho(\omega)$ of the velocity-autocorrelation function $\rho(t)$ is equal to $\lim_{k\to0}[2\pi\omega^2S_s(k,\omega)/k^2]$ and therefore behaves as[9]

$$\hat\rho(\omega)=2D-8\sqrt{2}\delta^{3/2}|D\omega|^{1/2}/(3k^*)+O(|\omega|^{3/4})$$

for small ω. The term proportional to $|\omega|^{1/2}$ in $\hat\rho(\omega)$ is due to the second term on the right-hand side of Eq. (2) [using Eq. (3)] and leads, after Fourier transformation, to the long-time tail $\sim t^{-3/2}$ in $\rho(t)$. Thus, Eq. (2) is consistent with a tail in $\rho(t)$ and therefore with the existence of mode-coupling effects in $D=\int_0^\infty dt\,\rho(t)$.

We will also consider the top value $S_s(k,0)$ of $S_s(k,\omega)$ and the half-width $\omega_H^s(k)$ which is defined by $S_s(k,\omega_H^s(k))=\frac{1}{2}S_s(k,0)$. In fact we study

$$\pi k^2S_s(k,0)=D^{-1}+D^{-1}G(\delta^{-1})k/k^*+O(k^{3/2})\ , \tag{4a}$$

$$\omega_H^s(k)/k^2=D-DH(\delta)k/k^*+O(k^{3/2})\ , \tag{4b}$$

$$\pi S_s(k,0)\omega_H^s(k)=1+A(\delta)k/k^*+O(k^{3/2})\ , \tag{4c}$$

where

$$H(\delta)=1.4531\delta^{3/2}[1-0.7276\delta-0.1523\delta^2+O(\delta^3)]\ ,$$

$$A(\delta)=1.2136\delta^{3/2}[1+0.212\delta+0.065\delta^2+O(\delta^3)]$$

and where G, H, and A are positive for all physical values of δ (i.e., $0\le\delta\le1$). Since in Eqs. (4) the terms linear in k arise from the leading mode-coupling contribution in Eq. (2), these terms are the analogs of the long-time tail in $\rho(t)$. For the present sample of H_2 the experimental value[10] for $\nu=17.05$ Å^2/ps. We could not find an independently obtained experimental value for D. There are, however, a number of semiempirical procedures to estimate D.[18–20] Of these we use the following three.

First, for simple fluids like Ar, Kr, Xe, and CH_4 the actual values of D are within 10–20 % equal to the values of $D_{\rm HS}$ obtained for hard-sphere systems at the same temperature and density and with the same mass of the particles. Thereby the corresponding equivalent hard-sphere diameter $\sigma_{\rm HS}$ has been determined using different approaches[20–22] with slightly different results for $\sigma_{\rm HS}$. From the effective pair potential of hydrogen,[23] we estimate for H_2 at the present thermodynamic state that 2.70 Å $\le\sigma_{\rm HS}\le$ 2.85 Å so that $0.42\le n\sigma_{\rm HS}^3\le0.49$ and therefore[5,20] 6.7 Å^2/ps $\ge D_{\rm HS}\ge$ 5.2 Å^2/ps.

Second, a study of the Stokes-Einstein relation between D and ν revealed that for dense gaseous methane[20] $\beta MnD\nu=(0.125\pm0.005)\sigma_{\rm HS}^{-1}$ for $0.4\lesssim n\sigma_{\rm HS}^3\lesssim0.5$. Using

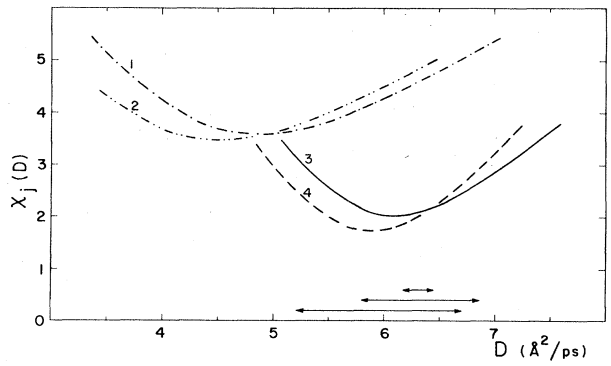

FIG. 1. The root-mean-square relative deviations $\chi_j(D)$ of the experimental neutron spectra for hydrogen at 120 K and 730 bar from two theoretical predictions, as functions of the self-diffusion coefficient D. The curves $j=1,2$ and 3,4 are the deviations from the simple diffusion law and mode-coupling theory, respectively [cf. Eqs. (1)–(3)]. The curves $j=1,3$ and 2,4 involve all experimental data points with 0.42 Å$^{-1}\le k\le$ 0.84 Å^{-1} and 0.42 Å$^{-1}\le k\le$ 1.05 Å^{-1}, respectively. The horizontal arrows represent three independent estimates for D. Note the minimum in χ_3 at $D=6.1$ Å^2/ps.

this relation for H_2 yields for the present case $D_{SE}=6.3\pm0.5$ Å^2/ps. We remark that for methane in the same density range, values for ν/ν_E are obtained which are equal to 0.90 ± 0.05, where ν_E is Enskog's value for ν of the equivalent hard-sphere gas. Such values of ν/ν_E are obtained here for H_2 when $\sigma_{HS}=2.70\pm0.05$ Å, consistent with the estimate for σ_{HS} given above.

Third, experimental data for D agree within about 10% with those obtained from molecular dynamics (MD) results for equivalent Lennard-Jones (LJ) fluids.[18] A recent MD experiment for a LJ fluid of 864 particles with masses equal to those of H_2 molecules, at the present density and temperature of the H_2 sample and with[24] $\epsilon_{LJ}/k_B=36.7$ K and $\sigma_{LJ}=2.96$ Å yields $D_{LJ}=6.30\pm0.15$ Å^2/ps.[25] The three estimates for D obtained here are represented in Fig. 1 by horizontal arrows. We remark that they are consistent with one another.

IV. RESULTS

In order to investigate to what extent experimental neutron spectra $I_c(\varphi,\lambda)$ are described by the Eqs. (1) and (2), in how far the mode-coupling corrections in Eq. (2) are relevant and up to which k values, we proceed as follows. First we determine the root-mean-square relative deviations $\chi_j(D)$ of $I_c(\varphi,\lambda)$ from the theoretical predictions [cf. Eqs. (1) and (2)] as functions of D for four cases j. Here

$$\chi_j(D)=\left[(N_j-1)^{-1}\sum_{l=1}^{N_j}\Delta_l^2/s_l^2\right]^{1/2},$$

where l runs over all N_j discrete experimental data points of $I_c(\varphi,\lambda)$ which are involved in case j, s_l is the estimated standard deviation of the experimental error, and Δ_l is the difference between $I_c(\varphi,\lambda)$ and the theoretical prediction. The cases $j=1$ and 2 involve all data points of $I_c(\varphi,\lambda)$ with $k(\varphi,\lambda)\leq0.84$ Å^{-1} and $k(\varphi,\lambda)\leq1.05$ Å^{-1}, respectively, and the theoretical prediction given by the first term on the right-hand side of Eq. (2) only. The cases $j=3$ and 4 involve the data points with $k\leq0.84$ Å^{-1} and 1.05 Å^{-1}, respectively, and the theoretical prediction *including* the mode-coupling term in Eq. (2). In these fits $N_1=N_3=695$, $N_2=N_4=1400$, and the smallest value of $k(\varphi,\lambda)$ obtained in the experiment is 0.42 Å^{-1}. The results for $\chi_j(D)$ are displayed in Fig. 1. We find for the present neutron spectra that values of χ equal to 3, 2, and 1 correspond to fits in which, respectively, 50%, 15%, and 2% of the data points deviate more than their estimated standard deviation from the theoretical predictions and that the points which deviate most are concentrated near λ_0. Thus we find that our experimental results for $I_c(\varphi,\lambda)$ are very poorly described by Fick's law at any value of D (cf. Fig. 1) both for 0.42 Å$^{-1}\leq k\leq0.85$ Å^{-1} ($j=1$) and 0.42 Å$^{-1}\leq k\leq1.05$ Å^{-1} ($j=2$). We see in Fig. 1 that $I_c(\varphi,\lambda)$ is far better represented by the prediction from the mode-coupling theory [cf. Eq. (2)]. The best fits are obtained for $D=6.1$ Å^2/ps ($j=3$) and 5.9 Å^2/ps ($j=4$), respectively. Note that these values are in the range estimated for D (cf. Fig. 1).

Next we display in Fig. 2 the experimental results for $I_c(\varphi,\lambda)$ at six different angles φ as functions of λ. These are representative for all 15 spectra. Also shown are the full widths at half maximum of the resolution functions $R(\varphi,\lambda'')$. We note that the $R(\varphi,\lambda'')$ are fairly well represented by normalized Gaussians with full widths equal to 0.061 Å for all φ. Furthermore in Fig. 2 we display the predictions by Fick's law and the mode-coupling theory [cf. Eqs. (1) and (2)] with $D=6.1$ Å^2/ps for all φ.

Finally, we determine $S_s(k,0)$ and $\omega_H^s(k)$ from the experimental data points in the following way. We use a

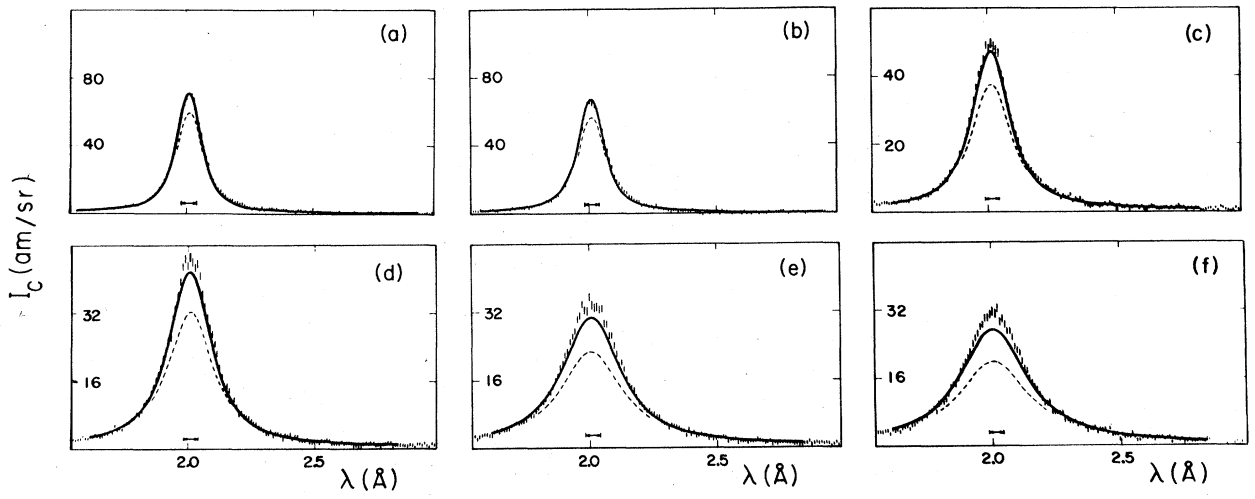

FIG. 2. Corrected experimental neutron spectra $I_c(\varphi,\lambda)$ for hydrogen (vertical bars) as functions of the wavelength λ of scattered neutrons at scattering angles $|\varphi|$: (a) 8.96° (b) 9.18°, (c) 12.02°, (d) 13.04°, (e) 15.82°, and (f) 17.04°. The wavelength λ_0 of incoming neutrons is 2.013 Å. The solid and dashed curves represent the predictions from the mode-coupling theory and the simple diffusion law, respectively, where, in all cases, $D=6.1$ Å^2/ps. The horizontal arrows represent the full widths at half maximum of the experimental resolution functions $R(\varphi,\lambda'')$.

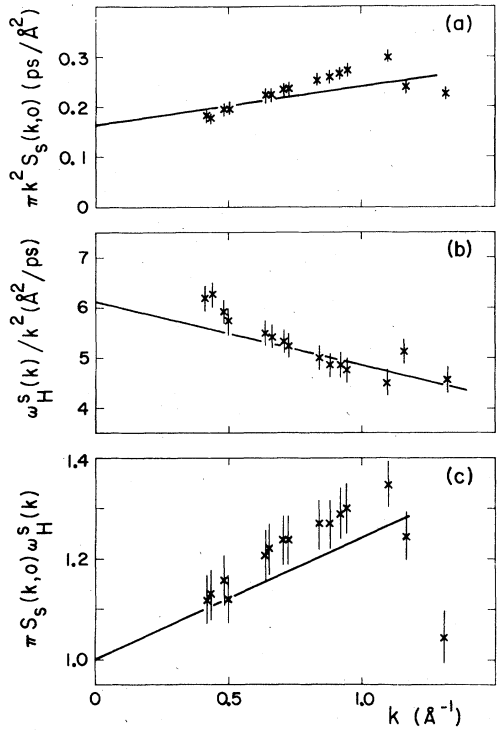

FIG. 3. Reduced top value $S_s(k,0)$ (a) and half-width $\omega_H^s(k)$ (b) of $S_s(k,\omega)$ and their product (c) as functions of k for hydrogen ($\times$) and according to the mode-coupling theory [cf. Eq. (4)] with $D=6.1$ Å^2/ps (solid lines).

representation for $S_s(k,\omega)$ which involves two adjustable parameters, the normalization factor N and the diffusion coefficient D. The explicit representation for $S_s(k,\omega)$ is given by the first two terms on the right-hand side of Eq. (2) both multiplied by N. We relate $S_s(k,\omega)$ to $I_c(\varphi,\lambda)$ using Eq. (1). From a least-squares-fitting procedure we determine for each φ the best $N(\varphi)$ and $D(\varphi)$ and the root-mean-square relative deviation $\chi(\varphi)$. For the smallest four angles φ we find very good fits [$\chi(\varphi)=1.3\pm0.2$], while for the remaining 11 angles the fits are perfect [$\chi(\varphi)=1\pm0.05$]. Using the explicit representation for $S_s(k,\omega)$ with the experimentally obtained values of $N(\varphi)$ and $D(\varphi)$ we determine at each φ, $S_s(k,0)$ and $\omega_H^s(k)$ numerically as functions of $k=k(\varphi,\lambda_0)$. The results are shown in Fig. 3. The uncertainties of about 10% in $S_s(k,0)$ and $\omega_H^s(k)$ are derived from the uncertainties in $I_c(\varphi,\lambda)$ at $\lambda\simeq\lambda_0$. Also shown are the mode-coupling predictions [cf. Eq. (4)] with $D=6.1$ Å^2/ps. We note that in the fits described above values of $N(\varphi)$ and $D(\varphi)$ are obtained which are equal to $N(\varphi)=1.03\pm0.03$ and $D(\varphi)=6.0\pm0.5$ Å^2/ps. These values are consistent with the uncertainty in the absolute normalization of $I_c(\varphi,\lambda)$ and with the independent estimates for D, respectively.

V. CONCLUSIONS AND DISCUSSION

We find (cf. Fig. 1) that the measured neutron spectra of dense hydrogen at 120 K and 730 bar cannot be described by Fick's law when $k\geq0.42$ Å^{-1}. For the k values considered here we find, in fact, that $S_s(k,\omega)$ cannot be described by any normalized Lorentzian function in ω. This follows from the fact that for normalized Lorentzians $\pi S_s(k,0)\omega_H^s(k)=1$, which value differs significantly from the present results for this quantity, as is shown in Fig. 3(c). However, a good overall description of our neutron spectra is obtained if we use the mode-coupling theory [cf. Eq. (2)] with values of D which are within the ranges estimated in three independent ways [cf. Figs. 1 and 3(c)]. In our view this implies that the mode-coupling theory might well be relevant for the description of neutron spectra at small angles. Furthermore we find [cf. Figs. 2(a)–2(c)] that the mode-coupling theory describes $I_c(\varphi,\lambda)$ very well for all λ in the range of scattering angles $|\varphi|$ between 9° and 12° and with $D=6.1$ Å^2/ps. We note, however, that for increasing $|\varphi|$ the theoretical prediction increasingly deviates from $I_c(\varphi,\lambda)$ at $\lambda\simeq\lambda_0$ [cf. Figs. 2(d)–2(f)]. We observe in Figs. 3(a) and 3(b) that $\pi k^2 S_s(k,0)$ increases and $\omega_H^s(k)$ decreases with increasing k. The behavior of both quantities is in reasonable agreement with the linear laws predicted by the mode-coupling theory [cf. Eq. (4)], although deviations are found for $S_s(k,0)$ at $k\sim1$ Å^{-1}. However, since we find no significant changes in $\chi_j(D)$ for the cases $j=3$ and 4 (cf. Fig. 1), we conclude from this and the foregoing that the mode-coupling theory is at least relevant up to wave numbers k of the order of 1 Å^{-1}.

Finally we discuss the following features of our present results.

(1) At each φ we find that our neutron spectra can be described by *unnormalized* Lorentzians, i.e., by the first term on the right-hand side of Eq. (2) multiplied by a normalization factor N. Then, reasonably good fits are obtained (i.e., $\chi\simeq1.5$) when $N\simeq1.25$. These values of N are far outside the present experimental uncertainty for the normalization of $I_c(\varphi,\lambda)$ however. Thus, also these fits are inconsistent with the predictions by Fick's law.

(2) For the k values considered here the corrections to Fick's law due to mode coupling are of the order of 20–30% (cf. Fig. 2). Remarkably enough, for hard spheres at the same density, the mode-coupling correction to Enskog's value of D is of the same order of magnitude.[5,6,20]

(3) It has been found before that the mode-coupling theory can account for the experimentally observed anomalous sound dispersion in liquid argon.[26] The range of k values for which theory and experiment agree is similar to that found here (i.e., $k\sigma_{HS}\lesssim3$) as is the order of magnitude of the mode-coupling corrections in both cases.

(4) For decreasing k the approach of $\omega_H^s(k)/k^2$ to D from *below* [cf. Fig. 3(b)] has been observed before in hydrogen,[27] liquid argon,[28] and sodium.[29] The mode-coupling theory [cf. Eq. 4(b)], might well give an explanation for this so-called diffusion retardation.[29]

(5) In the analysis of their neutron spectra Chen *et al.*[1] assumed the existence of a finite range of k values, from $k=0$ upwards, for which $\omega_H^s(k)/k^2$ is constant and therefore equal to D. Their measurements were consistent with such an assumption and thus values for D could be derived from the neutron spectra at a series of thermodynamic states. We find for all the cases discussed in Ref. 1 that the results for $\omega_H^s(k)/k^2$ are also consistent

with the mode-coupling theory, i.e., with the linear law given by Eq. 4(b). However, the values for D derived from these results on the basis of Eq. 4(b) are slightly larger than those obtained using the assumption made in Ref. 1. The agreement of these new values for D with those of the equivalent hard-sphere gases discussed in Ref. 1 is somewhat better than the agreement found by Chen *et al.* It seems, therefore, that the mode-coupling theory might be relevant also for the results for $\omega_H^s(k)/k^2$ reported in Ref. 1.

(6) From a theoretical calculation we find that the mode-coupling terms of order $k^{3/2}$ in Eq. (4) are at most 1% of the terms which are linear in k when $k \simeq 1$ Å^{-1}. Therefore the deviations of theory and experiment for $k \simeq 1$ Å^{-1} [cf. Figs. 3(a), 3(c), and 2(d)–2(f)] cannot be understood on the basis of these higher mode-coupling corrections. Most probably the deviations are due to kinetic effects[1] which are of order k^2 in Eq. (4). Thermodynamic states of H_2 for which these kinetic effects are expected to be smaller are presently under investigation. Such experiments, with higher accuracies and at smaller wave numbers than obtained in the present experiment, together with an independent and precise knowledge of D, might well yield convincing evidence for the existence of mode-coupling effects in dense hydrogen.

(7) It is not possible to obtain the Fourier transform $\hat{\rho}(\omega)$ of the velocity-autocorrelation function from our present experimental data. Since the data for $\omega^2 S(k,\omega)/k^2$ should be extrapolated to $k=0$, accurate measurements are needed in particular at small k. However we could not reliably correct the spectra at the smaller scattering angles for instrumental resolution. How small k should be in the new measurements to make an extrapolation to $k=0$ feasible is presently under investigation.

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