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Quasielastic neutron scattering measurements of fast local translational diffusion of lipid molecules in phospholipid bilayers

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Quasielastic incoherent neutron scattering has been used to investigate the rate of local translational diffusion of lipid molecules in phospholipid bilayers of dipalmitoyl-phosphatidylcholine. The measured translational diffusion constants ($4 \cdot 10^{-10}$ m^2 s^{-1} at 63°C and $1.4 \cdot 10^{-11}$ m^2 s^{-1} at 30°C) are considerably faster than those deduced using other less direct methods, but are in agreement with those measured in soap-water lyotropic liquid crystals, and with calculated values. This disagreement is attributed to differences in the time and distance scales characterising the various measurements. Quasielastic neutron scattering experiments observe fast motions over molecular distances, whereas other methods tend to measure a rate of diffusion which is averaged over macroscopic distances, and may thus contain contributions from long distance slow diffusive motions such as diffusion between the bilayers.

Introduction

An important property of water lipid mesophases is the molecular fluidity of the lipids. Because they are directly related to biological membranes considerable attention has been payed to phospholipid bilayers, and different aspects of their microdynamics have been studied [1–3]. One particularly important property is the rate of lateral diffusion (D) of the lipid within the bilayer. Although this is a quantity, which in heterogenous systems is not easily measured in a direct manner; for phospholipids in the liquid-crystal state, values of between 10^{-14} m^2 s^{-1} and 10^{-11} m^2 s^{-1} have been published [1–12]. This is significantly slower than the values of the diffusion constant found in bulk liquid paraffins [13] ($2.2 \cdot 10^{-9}$ m^2 s^{-1} for *n*-octane at 25°C; $4.6 \cdot 10^{-10}$ m^2 s^{-1} for *n*-octadecane at 50°C) and also of the measured rates of lipid diffusion in soap-water lyotropic liquid crystals [13–18], micelles, and microemulsions [19,20] (10^{-9}–10^{-10} m^2 s^{-1}). Moreover, calculations [21,22] predict rates of lateral diffusion in agreement with these values. It is thus suprising that the reported diffusion constants of the lipid in phospholipid bilayers are over two orders of magnitude slower than this. Although phospholipids contain two paraffinic chains, whereas most of the soap molecules whose diffusivity has been studied contain only one, this does not satisfactorily account for such a substantial slowing down of the lipid molecules. Pace and Chan [22] have calculated lateral translational diffusion constants in bilayers and predict only a 2- or 3-fold reduction in the diffusivity on going from a single to a double chain surfactant molecule. On this basis therefore, the rate of lateral diffusion in the liquid crystal phase of a phospolipid bilayer might be expected to be 10^{-10} m^2 s^{-2} rather than 10^{-12} m^2 s^{-1}.

The methods normally used to derive diffusion constants in lipidic layers, such as NMR [3–5,7–9], EPR [6], and fluorescence [10–12], tend to measure diffusion over distances ($> 10^4$ Å) and/or times ($> 10^{-9}$ s) which are relatively long. Incoherent quasielastic neutron scattering [23–27] (QENS) measures the motions of protons over distances (2–100 Å) and times ($< 10^{-9}$ s) which are much shorter, and correspond to those involved in molecular diffusion. QENS studies of lipidic diffusion in bulk paraffins [27], lyotropic liquid crystals [18], micelles [19] and microemulsions [20], give diffusion constants of between 10^{-9} m^2 s^{-1} and 10^{-10} m^2 s^{-1}. Preliminary QENS measurements on phospholipid layers have been made by Mittendorf and Stirling (personal communication). In the present paper we present

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QENS measurements of lipidic motions in bilayers of dipalmitoylphophatidylcholine (DPPC) in both the liquid crystal and gel phases. The values of the translational diffusion constant derived ($4 \cdot 10^{-10}$ m^2 s^{-1} at 63°C and $1.4 \cdot 10^{-11}$ m^2 s^{-1} at 30°C) are significantly faster than those reported using other more indirect methods, but are in agreement with the values anticipated using the arguments presented above.

Quasielastic neutron scattering

In these experiments [23–28] the sample is placed in a monochromatic neutron beam. As a result of molecular motion, energy is transferred between the neutrons and the sample, and the resulting energy broadening of the scattered neutrons is measured at different angles. In hydrogen containing systems the scattering is dominated by the incoherent contribution from the protons and the scattering can be interpreted in terms of the individual motions of the protons.

The width of the energy broadening is related to the time scale of the molecular motions which contribute to it, whereas the angular dependence of the broadening yields information about the type and spatial extent of these motions [23–28]. The angular dependence is expressed in terms of reciprocal space, ($Q = 4\pi/\lambda \sin\theta$ where 2θ is the scattering angle and λ the neutron wavelength). Fickian translational diffusion leads to a Lorentzian shaped broadening whose width increases linearly with Q^2, and a plot of broadening against Q^2 can be used to derive the translational diffusion constant. In contrast to this, rotational motions result in a spectrum comprised of a Lorentzian shaped quasielastic peak superimposed on an elastic component. The width of the quasielastic peak is independent of Q and gives the rotational diffusion constant, whilst the dependence of the intensities of the elastic and quasielastic components with Q is related to the spatial trajectory described by the rotating protons.

Simple and convincing evidence for fast translational diffusion is provided when the plot of broadening against Q^2 is linear and passes through the origin. Moreover, the values of the diffusion constants derived from such plots, are to within a factor of about two, independent of whether the diffusive motions are two- or three-dimensional in nature or if they are in some way restricted in space [24,28]. QENS has its own intrinsic time and distance scales which are approximately 10^{-12}–10^{-9} s and 10^0–10^3 Å. The 'observation' distance is related to the value of Q at which the measurement is made, and which varies with scattering angle.

The combination of the direct observation of fast motions over microscopic distances makes QENS a powerful method for the study of molecular dynamics. Furthermore, because the 'observation' distance is well defined and variable, and can nearly always be made to be smaller than the distance over which translational motions may be restricted, the method is well suited to the study of heterogeneous systems.

The incoherent scattering cross section of hydrogen is much greater than that of other atoms and is nearly 40-times greater than that of the deuteron. Because of this and because incoherent quasielastic spectra are the sum of the contributions from the individual protons, the spectrum of any component in a multicomponent system may be obtained by subtracting the spectrum of the completely deuterated system from that in which the selected component is protonated. For example, in a dispersion comprised of 20% phospholipid in D_2O, the incoherent scattering from the phospholipid molecules will be 5- to 10-times greater than that from D_2O. Hence subtraction of the D_2O spectrum from the phospholipid containing spectum will give the spectrum of the phospholipid on its own.

Materials and Experimental Details

Fully protonated DPPC (DPPC-h) was purchased from Sigma (St. Louis, U.S.A.) and used without further purification. DPPC in which the palmitoyl paraffinic chains had been selectively deuterated (DPPC-d) was prepared in the following manner. Glycerophospocholine-cadmium chloride complex (Calbiochem, San Diego, U.S.A.) was acylated with perdeuterated palmitic acid (CEA, France) in dimethylformamide using 4-dimethylaminopyridine as a catalyst [29], and purified by silicagel chromatography (Bio-Sil-1 from Bio-Rad Laboratories, Richmond, U.S.A). All lipid samples were found pure by thin-layer chromatography.

Samples were prepared by dissolving 0.50 g of DPPC-h and 0.54 g of DPPC-d in 2.5 ml each of D_2O. At these concentrations a lamellar structure is formed and this was checked by X-ray diffraction. At 55°C the samples were in a liquid crystalline state whereas at 30°C they were in the 'gel' phase; the transition between the two occurring at approx. 43° C.

QENS measurements were made using the IN5 and IN10 spectrometers at the Institut Laue-Langevin, Grenoble [30]. The IN5 measurements used incident neutron wavelengths of 6 Å, 8 Å and 10 Å and had spectral windows of 1700, 700 and 300 μeV respectively. The energy resolution as determined with a 1 mm vanadium plate was 40 μeV, 18 μeV, and 8 μeV half-width at half-maximum (HWHM) for the 6 Å, 8 Å and 10 Å measurements, respectively. The detectors were arranged so as to simultaneously measure energy spectra at seven different values of momentum transfer between 0.4 $Å^{-1}$ and 1.5 $Å^{-1}$. The IN10 measurements used an incident neutron wavelength of 6.28 Å. The energy resolution was 1.2 μeV HWHM, the spectral window was 15 μeV, and spectra were simultaneously

measured at seven different values of momentum transfer from 0.2 $Å^{-1}$ to 1.3 $Å^{-1}$.

The samples were contained in flat aluminium cells having a sample thickness of 1 mm and oriented at 135° to the neutron beam. They were maintained at the desired temperature by a water circulation bath and the temperature was measured with thermocouples inserted into the sample holder. Counting times per run varied from between 3 h and 36 h but were often of the order of 12 h. Some difficulty was experienced in initially adjusting the sample temperature and keeping it completely stable over the duration of the experiment (approx. 7 days per instrument). This accounts for the slightly different sample temperatures of the IN5 (63°C) and IN10 (55°C) measurements. In both cases the maximum deviation from the average temperature was ±3°C.

Data were corrected for detector efficiency and background scattering by means of standard procedures and programmes available at the I.L.L. No corrections were made for multiple scattering. The spectrum corresponding to the phospholipid was obtained by subtracting the spectrum from D_2O after correcting for the difference in transmissions and excluded volumes.

The resulting spectra were fitted to a convolution of the experimentally determined resolution function with one or more Lorentzian shaped broadenings and an elastic peak. This gave the width(s) and intensitie(s) of the Lorentzian broadening(s), and the intensity of the elastic component. Using this procedure all the spectra were satisfactorily described by a single Lorentzian broadening.

Results

QENS spectra were taken using the medium resolution instrument, IN5, for DPPC in the liquid crystal phase (63°C); and with the high resolution instrument, IN10, in both the liquid crystal (55°C) and gel phases (30°C). No IN5 measurements were made on the gel phase because the expected broadenings were less than the instrument resolution.

In all cases the deconvoluted half width of the energy broadening increased linearly with Q^2. This shows that the observed spectra result principally from translational diffusion of the phospholipidic protons. The Q range covered using the medium resolution instrument, IN5, was from 0.4 $Å^{-1}$ to 1.6 $Å^{-1}$, and this corresponds to 'observation' distances of from approx. 4 Å to 15 Å. Some of these distances are sufficiently small for it to be possible to attribute part of the observed effect to a librational motion of the two palmitoyl paraffinic chains, rather than to a translational displacement of the whole phospholipid molecule. To check against this possibility, two additional experiments were performed. The first consisted of making measurements at lower values

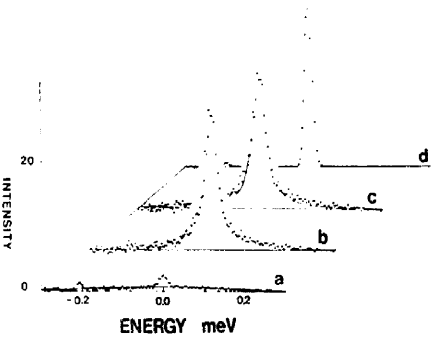

Fig. 1. Neutron quasielastic scattering spectra at 63°C measured on IN5 [30] using 10 Å wavelength neutrons and at a scattering vector of 0.92 $Å^{-1}$: (a) D_2O; (b) DPPC-h in D_2O; (c) DPPC-h spectrum obtained by subtracting (a) from (b); (d) vanadium (instrument resolution). In (d) the dotted line is to guide the eye between the points. In (c) the full line is a theoretical fit assuming a Lorentzian broadening. In (a) the full line indicates the broadening from D_2O; the elastic peak in (a) arises from scattering from the sample holder. The intensity is in arbitrary units.

of Q, and hence to observation distances which are greater than the possible displacement of a librating chain. If the resulting broadenings still increase linearly with Q^2 then the observed effect must arise not from librations, but from a molecular displacement. Because the energy broadening decreased rapidly with decreasing Q, these measurements required the high-resolution instrument IN10. The second experiment consisted of making measurements on DPPC in which the two palmitoyl paraffinic chains had been deuterated. Since the resulting spectrum will arise from the protons on the phospholipid polar headgroup only, and do not contain any contributions from the palmitoyl chains, any translational motion detected must correspond to a displacement of the polar head over the bilayer surface and cannot arise from librational motions of the paraffin chains.

It is convenient to separate the results into those obtained in IN5 at medium resolution (10–50 μeV) and those obtained on IN10 at high resolution (1 μeV). Fig. 1 shows the quasielastic spectrum measured from DPPC-h in D_2O at 63°C, compared with those measured under the same conditions from D_2O and a vanadium sample; the vanadium spectrum corresponds to the instrument resolution. As expected the spectrum from the phospholipid-containing system is much more intense than that from D_2O. Also shown in Fig. 1 is the spectrum corresponding to that from the phospholipid obtained by substracting the D_2O background spectrum from the phospholipid/D_2O spectrum. When subtracting the D_2O background spectrum, corrections were made for the difference in excluded volume and neutron transmissions of the two samples. However, these cor-

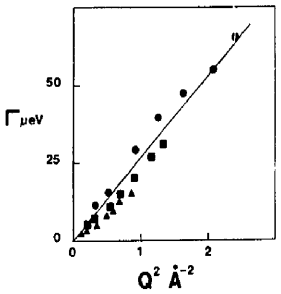

Fig. 2. Variation of the deconvoluted Lorentzian half-width (Γ) with Q^2 for DPPC-h at 63°C (IN5 measurements). Points of different shape are used for different incident neutron wavelengths: ●, 6 Å; ■, 8 Å; ▲, 10 Å. The error in the half-width depends upon the broadening and the resolution, but is approx. 15%. The straight line corresponds to a diffusion constant of $4.1 \cdot 10^{-10}$ m^2 s^{-1}.

TABLE I

Dependence of the deconvoluted half-width of the qausielastic broadening (Γ) on momentum transfer (Q), determined on IN5 using different neutron wavelengths, for DPPC-d at 63°C

The estimated error in the half-widths is ±15%. A plot of against Q^2 showed a linear behaviour whose slope gave a diffusion constant of $3.1 \cdot 10^{-10}$ m^2 s^{-1}.

6-Å neutrons		8-Å neutrons		10 Å neutrons	
Q (Å^{-1})	Γ (μeV)	Q (Å^{-1})	Γ (μeV)	Q (Å^{-1})	Γ (μeV)
0.59	10	0.45	4.0	0.36	1.7
0.72	14	0.54	6.0	0.43	2.6
0.97	22	0.73	9.2	0.58	5.1
1.12	27	0.84	14.0	0.68	7.5
1.28	34	0.96	18.0	0.76	9.7
1.43	39	1.07	21.0	0.85	10.0
1.54	43	1.16	24.0	0.93	15.0

rections were small and did not effect the subsequent analysis and interpretation.

The DPPC-h spectra are significantly broader than the instrument resolution but narrower than the D_2O background spectra. This simple result shows that the phospholipidic protons are undergoing fast diffusive motions which are somewhat slower than those present in bulk water. Moreover, the energy broadening increases rapidly with momentum transfer, and this shows that the observed spectra are dominated by translational diffusion. A plot of the deconvoluted half-width at half-height (Γ) against Q^2 yielded a good linear dependence. This is shown in Fig. 2 compared with a straight line corresponding to a rate of diffusion of $4.1 \cdot 10^{-10}$ m^2 s^{-1}. In Fig. 2 data from the measurements using different neutron wavelengths have been superimposed. With 6-Å neutrons the energy broadening at low values of momentum transfer was less than the instrument resolution. Longer wavelength neutrons give a higher energy resolution but they also decrease the values of Q and consequently the observed broadenings. Hence, so as to determine the energy broadening over as wide a Q range as possible measurements at three different neutron wavelengths were made.

Similar results were obtained from the sample in which the two palmitoyl paraffinic chains had been deuterated. The spectrum now arises from the protons on the polar headgroup only and does not contain any contributions from the the two palmitoyl chains. Although the spectrum is much weaker than that from the fully protonated sample no problems in the subtraction were encountered. The ratio of the intensities of the protonated and deuterated samples was found to be in good agreement with the ratio of the proton concentrations in the two samples. The observed energy broadenings (Table I) are similar to those measured on the fully protonated sample and plotting a straight line through the Γ-Q^2 plot yielded a diffusion coefficient of $(3 \pm 1) \cdot 10^{-10}$ m^2 s^{-1}. This result shows that the rate of translational motion observed is the same for both the polar head and the complete molecule, and that it therefore arises from a displacement of the whole molecule.

The line broadenings and diffusion constant ($(4 \pm 1) \cdot 10^{-10}$ m^2 s^{-1}) from DPPC-h are slightly greater than those from DPPC-d ($D = (3 \pm 1) \cdot 10^{-10}$ m^2 s^{-1}). This difference probably arises from experimental errors. A difference in sample temperature of 2–3°C would account for this effect and is the most likely explanation.

The spectra measured on IN10 are similar to those from IN5, and Table II shows the derived energy broadenings. Plots of broadening against Q^2 were linear and the slopes yielded diffusion constants for DPPC-h of $1.2 \cdot 10^{-10}$ m^2 s^{-1} and $1.4 \cdot 10^{-11}$ m^2 s^{-1} at 55°C and 30°C, respectively. As expected the diffusion constant determined at 55°C using IN10 is less than that de-

TABLE 2

Dependence of the deconvoluted half-width of the quasielastic broadening (Γ) on momentum transfer (Q), determined on IN10, for DPPC-h at 55°C and 30°C.

The estimated error in the half-width is ±15%. Plots of Γ against Q^2 showed a linear behaviour, the slopes of the lines gave diffusion constants of $1.2 \cdot 10^{-10}$ m^2 s^{-1} at 55°C and $1.4 \cdot 10^{-11}$ m^2 s^{-1} at 30°C.

Q (Å^{-1})	Γ (μeV)	
	55°C	30°C
0.25	0.6	0.14
0.36	1.1	0.24
0.45	1.3	0.20
0.52	2.3	0.38
0.65	3.3	0.36
0.88	6.2	0.83
1.28	13.0	1.30

termined at 63°C using IN5. In a plot of ln D against $1/T$, the measurements at 30°C, 55°C and 63°C fell on a straight line having an activation energy of 4.4 kcal mol^{-1}. For the IN10 measurements the lowest value of Q for which measurements were made was 0.2 $Å^{-1}$ and this corresponds to an 'observation' distance of approx. 30 Å. The fact that a linear Γ-Q^2 behaviour is observed over distances which are greater than the possible displacement of a librating chain, confirm that the observed effects result from a fast translational displacement of the whole phospholipid molecule.

Rotational motions give rise to a Q-independent broadening. A significant rotational contribution to the spectra would result in Γ-Q^2 plots which are not linear and do not pass through the origin. For both the IN5 and IN10 results, the Γ-Q^2 plots are linear and their extrapolation goes either though or close to the origin. This argues for a negligible rotational contribution to the observed spectra. Also, rotational motions give rise to an elastic spectral component. The experimental spectra were satisfactotily described by the convolution of the resolution function with a single Lorentzian broadening, and there is no indication of an appreciable elastic component. This also is consistent with an interpretation of the spectra in terms of diffusive motions dominated by translational processes.

When translational diffusion is continous the energy broadening increases linearly with Q^2. For translational diffusion by jumps, the linewidth against Q^2 plots bend down and show a maximum. For values of Q corresponding to distances greater than the jump distance, the linewidth increases linearly with Q^2, whilst for values of Q greater than this the linewidth decreases. For DPPC the plots of energy broadening against Q^2 do not show a maximum or a bending over. This is in keeping with neutron scattering results on bulk paraffins [27] and other lipid systems [18–20] and suggests that the mechanism of translational diffusion over the Q range studied is continous rather than by large jumps.

The fitting of a Lorentzian form to the observed energy broadening implies that the diffusion constants obtained from the Γ-Q^2 plots correspond to those for a three-dimensional diffusive motion. Since in bilayers, lipid motions will be two-dimensional, the values of D derived above should be corrected by multiplying by 3/2. A better procedure is to use the scattering law for 2D-translational diffusion to analyse the spectra [24]. Analysing the results this way, gave linear Γ-Q^2 plots corresponding to 2D-diffusion constants from DPPC-h at 63°C of $(4.6 \pm 1) \cdot 10^{-10}$ m^2 s^{-1} and DPPC-d of $(3.8 \pm 1) \cdot 10^{-10}$ m^2 s^{-1}. These values compare well with those derived using the simple 3D approach.

For 2D translation, the shape of the energy broadening is not a Lorentzian, being considerably more peaked close to the origin. However, if as is the case here, the energy broadening is less than three or four times the instrument resolution, then the convolution of the instrument resolution with the 2D spectral form is indistinguishable from the convolution of the instrument resolution with a Lorentzian. This accounts for the fact that even though the lipidic motions are almost certainly 2D, the observed energy broadenings can be satisfactorily fitted by a Lorentzian form.

Conclusion

The linear dependence of the observed energy broadening with Q^2 shows that fast molecular translational motions are present in the phospholipid bilayer. Evidence that these motions do not arise from librational motions of the palmitoyl chains is furnished by the experiment on selectively deuterated DPPC where the palmitoyl chains are not 'seen'. Moreover, the same rate of diffusion was deduced from experiments in which the 'observation' distance was extended to values larger than those expected for a librating chain. If motion perpendicular to the layer is taken as being slower than in the layer, then these results strongly suggest that the derived rates of translational diffusion correspond closely to the true rate of local lateral molecular diffusion in the bilayer. These values are in agreement with calculations and with measurements of the lipid mobility in soap based systems, but are considerably faster than those previously reported ($\approx 10^{-12}$ m^2 s^{-1}). For such a slow rate of diffusion the expected quasielastic broadening (0.13 μeV at $Q = 1$ $Å^{-1}$) would be considerably lower than the energy resolution of IN5 (10 μeV) and IN10 (1 μeV) and the neutron quasielastic spectra should hence show an 'elastic' peak superimposed on a quasielastic component. The quasielastic broadening would give the rate of diffusion of the fast motion, whilst the elastic peak would arise from the unresolved slow motion. None of the spectra from either IN5 or IN10 show an appreciable elastic peak; hence the neutron measurements reported here show no manifestation of the slow motions deduced from other experiments. This is not to say that slow translational diffusion does not occur, but that it is an additional process to the local translational diffusion observed by the neutron experiments. If neutron measurements could be extended to much smaller values of Q (longer distances) then the slope of the Γ-Q^2 plots should eventually decrease until the diffusion constant became the same as that measured by the more macroscopic measurements.

The time and distance scales inherent to the neutron method correspond to those of fast molecular diffusion. Most other methods measure diffusion over a longer time scale. If molecular diffusion is limited to a heterogeneity smaller than the molecular displacement during this time ($2 \cdot 10^4$ Å for a diffusion constant of 10^{-9} m^2 s^{-1} in 10^{-3} s), then the real rate of molecular diffusion

may be underestimated. The rate of molecular diffusion may also be underestimated if additional slow motions, such as diffusion between heterogeneties, are present. With some methods complications in interpreation can arise. For example NMR relaxation times are strongly effected by reducing the dimensionality of the movement from three to two, and by restricting the spatial extent of the diffusion [31]. If these factors are not accounted for, then the derived rate of molecular diffusion may be two or three orders of magnitude too slow.

The disagreement between the neutron results and those from other methods can be attributed to the different time and distance scales involved. In our opinion the QENS experiments measure the local rate of molecular translational diffusion in the bilayer, whereas other measurements tend to measure a rate of diffusion which is averaged over macroscopic distances and may contain contributions from slow long distance diffusive motions such as diffusion between the bilayers.

In biology it is not so much the exact rate of molecular diffusion of the lipid in the membrane that is of importance, but the fact that the membrane is locally liquid. The results reported here confirm this, and suggest that the local rate of translational diffusion is much closer to the values found in bulk paraffins than hitherto thought.

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