

THz dynamics of hydrogen sulfide: Search for collective modes of a non-hydrogen-bonded analog of water

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Understanding the atomic-scale dynamics of liquids in the terahertz (THz) regime remains challenging because liquids sustain phonon-like collective excitations governed by complex interactions. To clarify the role of hydrogen bonding in such dynamics, we performed inelastic neutron scattering on hydrogen sulfide (H_2S) and its deuterated form (D_2S), molecular analogues of water without a hydrogen-bonded network. By combining coherent and incoherent measurements, we show that liquid H_2S exhibits a clear fast-sound mode, with a propagation velocity about twice the adiabatic sound speed, as in water. In contrast to hydrogen-bonded liquids, which display a secondary, nearly momentum-independent mode at 6–7 meV, no corresponding feature is observed in liquid D_2S . This absence highlights the central role of hydrogen bonding in enabling additional low-energy excitations. Overall, while fast sound is not exclusive to hydrogen-bonded systems, the detailed vibrational spectrum is strongly shaped by the local bonding environment.

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I. INTRODUCTION

The study of the complex dynamics of many-particle systems is relevant in a broad range of contexts, from exotic systems such as quark-gluon plasmas, which require the introduction of numerous quasiparticles across a wide energy range [1], to electron-driven collective atomic dynamics in liquid metals [2–10].

Nontrivial atomic-scale dynamics is observed in glasses [11–14] and in many classes of liquids. Particular attention has been devoted to hydrogen-bonded (HB) systems, beginning with the fundamental case of bulk water [15–21]. Extensive investigations have subsequently focused on water-based systems, including biological environments [22–31], aqueous solutions [32], nanoparticle suspensions [33–38], and confined water [39]. However, while in liquid metals the long-range Coulomb interaction provides a clear microscopic basis for the existence of well-defined collective modes [5–7], no

equally direct link exists between hydrogen bonding and the collective dynamics in HB liquids.

A key signature of HB dynamics is the presence of a dispersive high-energy mode, i.e., a longitudinal excitation propagating at a velocity c_∞ much larger than the hydrodynamic sound speed c_0 . This phenomenon was first observed in molecular dynamics simulations of water [40] and later confirmed by inelastic neutron and x-ray scattering (INS and IXS, respectively) [15–19]. It is commonly referred to as fast sound, although this term was originally introduced in the context of binary systems with large mass disparity, where it describes the excitations of the lighter component [41]. In the present context, the term does not imply this propagation mechanism, but is used more generally to denote a high-frequency dispersive mode with velocity exceeding the hydrodynamic sound speed, as is common in the literature on liquid water [42]. A similar behavior, with $c_\infty/c_0 \gtrsim 2$, has been reported in many hydrogen-bonded systems [22,25,26,43–45].

The large c_∞/c_0 ratio typical of HB liquids often remains substantial over a broad momentum range. Recently, similarly elevated values, $c_\infty/c_0 \simeq 1.64 \pm 0.10$ and $c_\infty/c_0 \simeq 1.50 \pm 0.04$, have been reported for acetone [46] and benzene [47], respectively. In contrast, in these cases, such pronounced values occur only in a narrow region around $Q \simeq 0.4 \text{ \AA}^{-1}$. These findings may be compared with those for the weakly bonded van der Waals liquid argon [48], where $c_\infty/c_0 \simeq 1.37 \pm 0.12$.

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For liquid metals (Li, Na, K, Rb, Cs, Al, Si, Ge, Ga, Sn, Cd, Hg, Pb, Bi, Cu) [6], typical values yield an average ratio $c_\infty/c_0 \simeq 1.16 \pm 0.12$, consistent with those reported for Fe and Cu [49] and Zn [49,50]. For molecular systems with complex bonding, sulfur provides an illustrative example: in liquid sulfur [51] (melting temperature $T_m \simeq 386$ K), measurements at $Q(0.5 \text{ \AA}^{-1})$ yield $c_\infty/c_0 = 1.463 \pm 0.028$ at $T = 413$ K and $c_\infty/c_0 = 1.418 \pm 0.091$ at $T = 453$ K.

Liquid water also hosts an additional, weakly dispersive excitation at $\simeq 5\text{--}7$ meV, observed for nearly a century [52] and attributed to O–O–O bending modes [53]. INS and IXS measurements show that this mode couples to both longitudinal and transverse dynamics [16,18,45], which has stimulated a growing interest in transverse-like excitations in liquids [54–58].

The hydrogen-projected vibrational density of states (DoS) of water, derived from incoherent neutron scattering [15,17,59–61], exhibits a pronounced peak in the same energy region, supporting the existence of this second mode. Neutron polarization analysis and molecular dynamics simulations have provided further insight into the relationship between coherent and incoherent dynamics [62]. Despite extensive work, the microscopic role of HB interactions in shaping these modes remains partially unresolved. For this reason, studying a molecular analog of water that lacks hydrogen bonding—such as hydrogen sulfide—offers a unique opportunity to isolate which dynamical features are directly linked to the presence of a hydrogen-bond network.

INS is ideally suited for this purpose because of the strong isotope contrast between hydrogen and deuterium, with hydrogen dominated by incoherent scattering ($\sigma_{\text{inc}} = 80.26$ barn, while $\sigma_{\text{coh}} = 1.76$ barn) and deuterium by coherent scattering ($\sigma_{\text{coh}} = 5.59$ barn, while $\sigma_{\text{inc}} = 2.05$ barn) [63]. We therefore performed a combined INS study of hydrogen sulfide (H_2S) and deuterium sulfide (D_2S).

Although oxygen and sulfur are chemically similar, their interaction with hydrogen differs significantly: water forms a strong HB network, whereas H_2S does not, due to sulfur's lower electronegativity. The S–D bond length ($1.336 \text{ \AA}$) is much longer than O–D in water ($0.958 \text{ \AA}$), reflecting the larger covalent radius of sulfur ($R_S = 1.05 \text{ \AA}$) compared to oxygen ($R_O = 0.66 \text{ \AA}$) [64]. Hydrogen sulfide is common in geochemical and astrophysical environments [65] and exhibits remarkable conductive and superconductive behavior under extreme pressures [66–71]. However, phonon-dispersion measurements are not available for its liquid phases. Previous work has only reported the hydrogen-projected DoS in selected solid phases [72].

The present experiment on the THz collective dynamics of H_2S and D_2S was carried out in the liquid phase, in thermal equilibrium with the coexisting vapour, at $T_{\text{exp}} = 205$ K. This choice ensures that the reduced temperature, $T_r = T_{\text{exp}}/T_t$, matches that of water at room temperature. For hydrogen sulfide, the triple-point temperature is $T_t = 187.6$ K [73]. At similar reduced temperature, the adiabatic sound velocities are remarkably close: 1347.2 m/s at 295 K for H_2O , and 1487.7 m/s at 205 K for H_2S [74].

The sample was measured at a pressure of approximately 0.067 MPa [74]. A strict scaling of the pressure with respect to the triple point would correspond to a pressure of about 4 MPa

at 205 K. While such a condition was not experimentally implemented, the associated change in density, and hence its effect on the collective dynamics, is expected to be negligible.

Interestingly, comparable values of c_0 are found in many molecular liquids, suggesting that sound velocity is not strongly influenced by hydrogen-bond networks. In the following, we show that despite this similarity in hydrodynamic sound speed, the THz dynamics of hydrogen sulfide differs markedly from that of water, revealing which features of water's spectrum specifically require hydrogen bonding.

II. EXPERIMENT AND DATA REDUCTION

The experiment on H_2S and D_2S was performed using the purpose-built BRISP time-of-flight spectrometer installed at the Institut Laue Langevin (Grenoble, France) [75,76]. Two experimental configurations were adopted, both exploiting a neutron beam with a fixed incident energy E_0 selected by a HOPG monochromator. The first configuration (004 reflection) selects neutrons with $E_0 = 83.7$ meV (corresponding to an incident neutron wavelength $\lambda \approx 1 \text{ \AA}$), while the second (002 reflection) provides $E_0 = 20.9$ meV ($\lambda \approx 2 \text{ \AA}$). The sample-to-detector distance was set to 4 m, yielding two different energy resolutions: a medium resolution at $\lambda = 1 \text{ \AA}$ and a higher resolution at $\lambda = 2 \text{ \AA}$. The BRISP sharp two-dimensional collimation (0.5°) and the fine spatial-resolution position-sensitive detector ensure a good angular and momentum resolution.

The two configurations cover the kinematic regions shown in Fig. 1: a broader dynamic range is obtained with $\lambda = 1 \text{ \AA}$, whereas $\lambda = 2 \text{ \AA}$ yields a narrower dynamic window but significantly improved energy resolution, allowing for a clearer separation of quasielastic and inelastic components of the dynamic structure factor. This complementary use of the two neutron wavelengths is essential: the $1 \text{ \AA}$ setting captures the full dispersion of the collective mode, while the $2 \text{ \AA}$ configuration resolves the narrow central peak and reveals subtle changes in the inelastic tail. The energy resolution function was determined using a vanadium standard. It is nearly Gaussian, with a FWHM of 2.5 meV ($E_0 = 83.7$ meV) and 0.56 meV ($E_0 = 20.9$ meV). The corresponding momentum resolutions were $0.07 \text{ \AA}^{-1}$ and $0.035 \text{ \AA}^{-1}$.

The sample was contained in a titanium slab-shaped container with 2-mm -thick windows, ensuring negligible deformation under the experimental conditions ($T_{\text{exp}} = 205$ K, $P_{\text{exp}} = 0.067$ MPa) and safe handling during sample loading at room temperature ($P \approx 2$ MPa) [74]. Standard ancillary measurements of the empty cell and of a full neutron absorber were carried out to evaluate the background. The sample transmission measurements agreed well with values expected from tabulated neutron cross sections [63]. The moderate energy dependence of the transmission for scattered neutrons was assumed to be identical to that of water, since no experimental data exist for H_2S or D_2S [77].

When subtracting the empty-cell signal, we assumed that in the explored (Q, ω) region, the titanium container produces only a small incoherent elastic contribution, as no Bragg peaks are present and lattice dynamics effects are weak. When the sample is present, the upstream window contributes an elastic signal attenuated by the sample, whereas the downstream

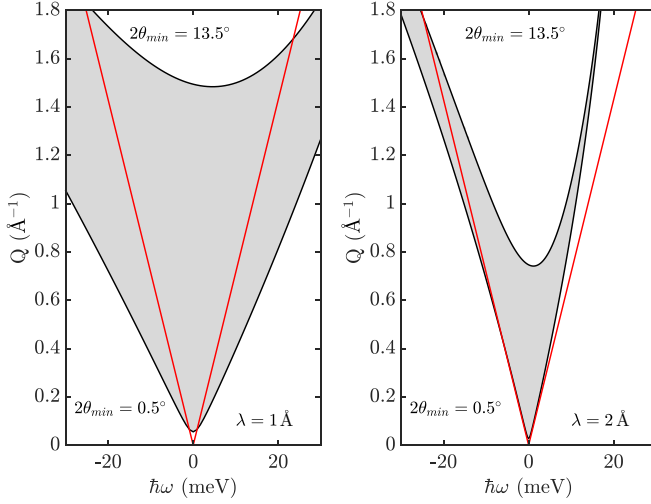


FIG. 1. Kinematical range covered by BRISP with the two experimental setups (gray area). The black lines indicate the Q - $\hbar\omega$ loci accessed at the minimum (0.5°) and maximum (13.5°) scattering angles 2θ . Left panel: $E_0 = 83.7$ meV. The elastic energy and momentum resolutions are $\Delta E = 2.5$ meV and $\Delta Q = 0.07 \text{ \AA}^{-1}$, respectively. Right panel: $E_0 = 20.9$ meV. The elastic energy and momentum resolutions are $\Delta E = 0.56$ meV and $\Delta Q = 0.035 \text{ \AA}^{-1}$. The two kinematical windows are shown on the same scale to highlight the different coverage. The straight red lines indicate the linear dispersion of an excitation propagating with velocity c_∞ , corresponding approximately to the position of the side-peak maxima extracted from the experimental data (see text).

window scatters at the energy of the exiting neutrons. A proper evaluation of the cell contributions was therefore performed, including the energy dependence of the Ti cross section. Multiple scattering effects were also estimated; owing to the relatively low molecular density, such contributions are small and smooth. The same modeling used previously for water [17] was adopted.

After applying background and multiple-scattering corrections, the resulting intensities were proportional to the differential scattering cross section for both D_2S and H_2S in the two configurations. Finally, the intensities were converted to absolute units using vanadium as a calibration standard.

III. RESULTS AND DISCUSSION

Representative results for the scattering intensities for D_2S and H_2S are shown in Figs. 2 and 3 for $\lambda = 1 \text{ \AA}$ and $\lambda = 2 \text{ \AA}$, respectively. The experimental data (black dots) are presented together with the instrument resolution determined from a vanadium standard (dashed line). For the D_2S sample, the fitting function is also shown along with its inelastic component $S_{in}(Q, \omega)$ (blue line), as discussed below. A markedly higher overall intensity is observed in H_2S (right panels), reflecting the dominant incoherent proton contribution.

Before proceeding with a quantitative analysis, it is useful to highlight the qualitative differences between the two samples. In D_2S , moderate but clearly distinguishable inelastic features are visible because the coherent signal is not fully masked by the incoherent quasielastic component. In contrast, H_2S —being dominated by hydrogen’s large incoherent

scattering cross section—shows a strong quasielastic signal that overwhelms the entire energy range.

This behavior is consistent with the known static structure factor of D_2S [78]. In the explored Q range (0.25 – $1.4 \text{ \AA}^{-1}$), the incoherent fraction is about 50% at low Q , decreasing to about 25% near the first diffraction maximum. For H_2S , the signal is overwhelmingly dominated by the proton incoherent cross section, with only a minor contribution from sulfur in both isotopic samples.

A significant part of the quasielastic peak arises from incoherent self-diffusion, modeled as a Lorentzian with FWHM $2\Gamma_d = 2DQ^2$, where $D = (3.7 \pm 0.2) \times 10^{-9} \text{ m}^2/\text{s}$ is the diffusion constant of H_2S [79]. For D_2S , the width scales approximately as the inverse square root of the molecular mass. This implies an FWHM narrower than the instrumental resolution for both systems: for example, $2\Gamma_d \approx 0.49$ meV for H_2S , and ≈ 0.34 meV for D_2S at $Q = 1 \text{ \AA}^{-1}$.

As observed in water and several other liquids, the dynamic structure factor of D_2S exhibits a pronounced and relatively smooth inelastic contribution (especially at $\lambda = 1 \text{ \AA}$), in addition to a resolution-limited central peak. These qualitative observations motivate the modeling approach discussed in the following subsection, where we separate quasielastic and inelastic components and determine the dispersion of the collective mode in D_2S .

A. Collective dynamics and dispersion curve in D_2S

To describe the D_2S spectra in Figs. 2 and 3, we used a minimal model with clear physical content, consisting of: (i) a quasielastic peak composed of two Lorentzian contributions (one associated with molecular self-diffusion and one broader term describing additional relaxational dynamics); and (ii) an inelastic component corresponding to a dispersive collective excitation. This modeling strategy mirrors that used for heavy water [15,17], ensuring that any differences between the two systems originate from the dynamics themselves rather than from the analysis procedure.

The need for two quasielastic components is clearly evident in the $\lambda = 2 \text{ \AA}$ data (left panel of Fig. 3), where the improved energy resolution allows one to distinguish the narrow central peak from the side features. Thus, the microscopic dynamics of D_2S is described by

$$S_{D_2S}(Q, \omega) = B(\omega)[A_1(Q)S_{qe1}(Q, \omega) + A_2(Q)S_{qe2}(Q, \omega) + S_{in}(Q, \omega)]. \quad (1)$$

Here, S_{qe1} and S_{qe2} are Lorentzian quasielastic components with Q -dependent amplitudes $A_1(Q)$ and $A_2(Q)$, and $B(\omega) = \beta\hbar\omega/(1 - e^{-\beta\hbar\omega})$ ensures detailed balance, with $\beta = 1/(k_B T)$.

The inelastic contribution is modeled using a damped harmonic oscillator (DHO) response,

$$S_{in}(Q, \omega) = A_{in}(Q) \frac{\Gamma(Q)}{[\omega^2 - \omega_c^2(Q)]^2 + [\Gamma(Q)\omega]^2}, \quad (2)$$

where $\omega_c(Q)$ is the mode frequency and $\Gamma(Q)$ is its damping. The DHO model captures the essential physics of longitudinal acoustic-like excitations in disordered systems, providing a

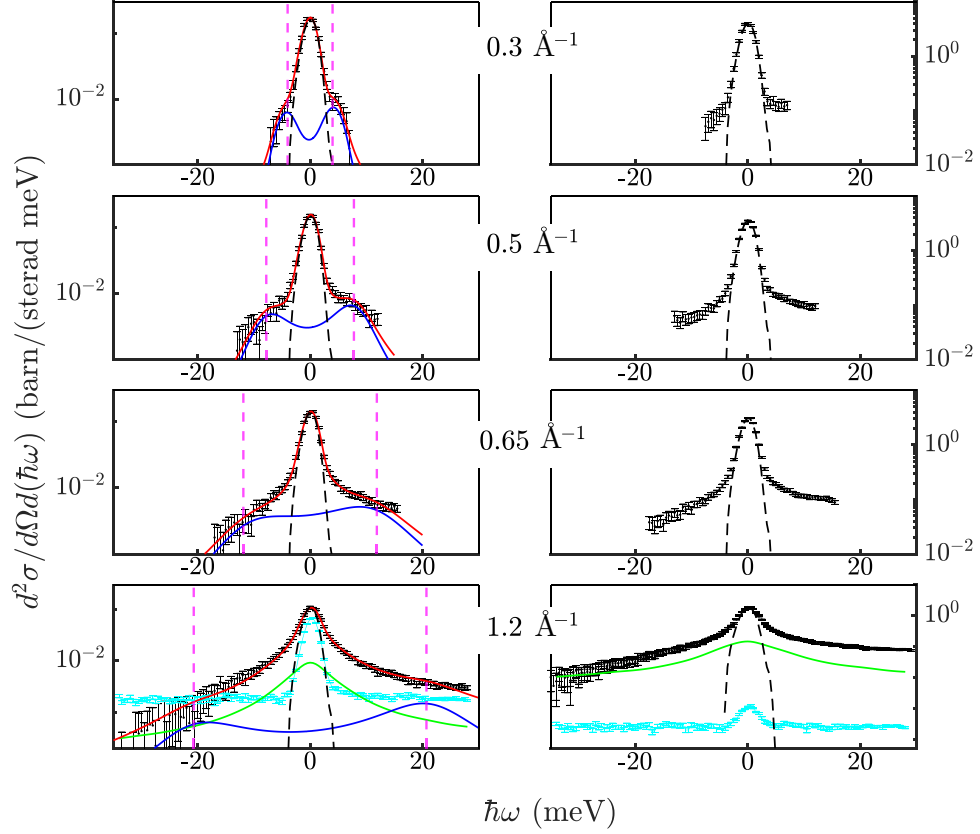


FIG. 2. Intensity as a function of energy transfer at selected Q values for D_2S (left) and H_2S (right), measured with $E_0 = 83.7$ meV ($\lambda = 1$ Å). The resolution function is shown as a dashed line. For D_2S , the red line is the fit using the model of Eq. (1), and the blue line is the corresponding inelastic component [Eq. (2)]. The background (cyan) and multiple scattering (green) contributions are also shown for the spectrum at $Q = 1.2$ Å $^{-1}$. The vertical magenta dashed lines indicate the excitation energies $\omega_c(Q)$ obtained from the model in Eq. (2) and shown in Fig. 4.

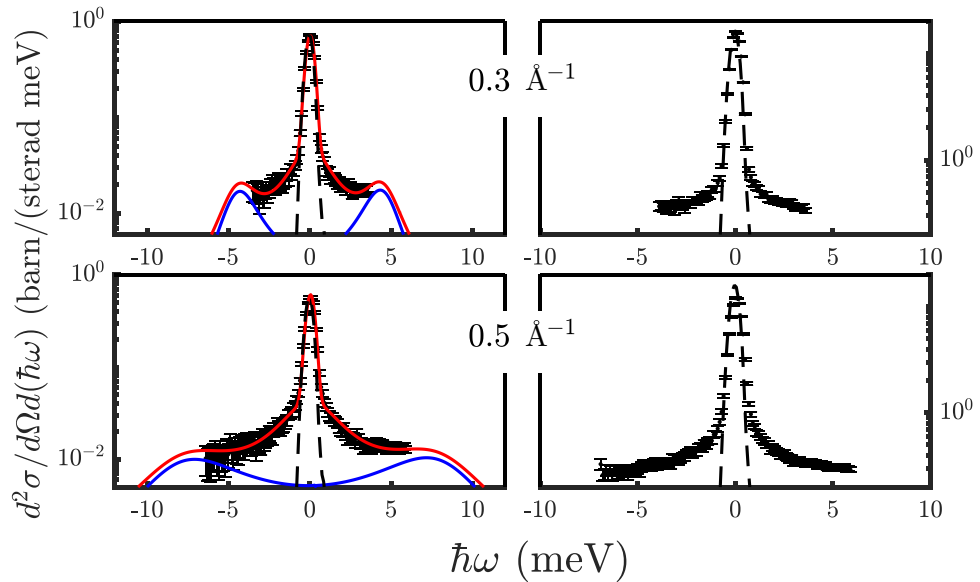


FIG. 3. Intensity versus energy transfer at two intermediate selected values of Q for D_2S (left) and H_2S (right), measured with $E_0 = 20.9$ meV ($\lambda = 2$ Å). The symbols follow the same convention as in Fig. 2.

robust estimate of both the dispersion and the damping of the mode.

For each Q value, the spectra measured at low and high energy resolution were fitted simultaneously using the same model function and a shared set of fit parameters. The model was separately convolved with the respective instrumental resolution functions of the two configurations. We note that the amplitude $A_{\text{in}}(Q)$ absorbs the normalization factors, ensuring dimensional consistency of $S(Q, \omega)$.

The resulting fits for both wavelengths are shown in Figs. 2 and 3. Two key points emerge:

- (i) The same model provides a consistent description of the spectra at both incident energies, requiring only a global resolution parameter; and
- (ii) the $\lambda = 2 \text{ \AA}$ data, owing to their superior (four times better) energy resolution, enable a much clearer separation between quasielastic and inelastic contributions.

As expected, the sharp quasielastic component corresponds to self-diffusion, giving a Lorentzian width $\Gamma_d \simeq DQ^2$ up to $Q \simeq 1 \text{ \AA}^{-1}$, the maximum momentum at which high-resolution data are available. The broader quasielastic term parallels that observed in heavy water [17], and it likely accounts for additional relaxational dynamics such as heat diffusion.

A well-known limitation of this type of nonlinear fitting is the strong correlation among $\omega_c(Q)$, $\Gamma(Q)$, and $A_{\text{in}}(Q)$ —especially at high Q , where both frequency and damping increase. Although these correlations affect the precision of the extracted parameters at large Q , the dispersion in the low- and intermediate- Q regions remains reliable, and provides the essential information on the presence (or absence) of secondary excitations. However, even an extended dynamic range would not remove this limitation, as additional high-energy modes—well known in the solid—would further complicate the spectra.

The D_2S data (left panels of Figs. 2 and 3) unequivocally reveal a single dispersing excitation. Extensive fitting attempts do not support the existence of a second weakly dispersive mode similar to the 5–7 meV mode characteristic of water. The resulting excitation frequencies $\omega_c(Q)$ and dampings $\Gamma(Q)$ are shown in Fig. 4. The dispersion also reports the adiabatic sound velocity of D_2S at 205 K, $c_0 = 1360 \text{ m/s}$ (8.945 meV $\text{\AA}$) [74]. As in water, the slope of $\hbar\omega_c(Q)$ is much larger than c_0 , demonstrating a clear fast-sound regime. A linear fit at $Q < 0.9 \text{ \AA}^{-1}$ yields $c_\infty = 2980 \pm 400 \text{ m/s}$ (19.6 ± 2.6 meV $\text{\AA}$), and thus $c_\infty/c_0 = 2.19 \pm 0.29$, comparable to values reported for hydrogen-bonded liquids.

The close similarity of c_∞/c_0 between D_2S and hydrogen-bonded systems is a central result of this study: it demonstrates that fast sound does not require hydrogen bonding and arises instead from more general features of liquid structure and interactions.

The present data demonstrate that fast sound ($c_\infty/c_0 > 1$) occurs systematically in the momentum range $0.2\text{--}0.3 \text{ \AA}^{-1}$ up to a finite fraction of the first diffraction peak. Hydrogen-bonded systems exhibit $c_\infty/c_0 \approx 2$ and a distinct secondary mode. Metallic liquids generally show $c_\infty/c_0 \approx 1$, except for Ca and Ba, whereas only weak hints of secondary modes have been reported for Na [80], Ga [81], and Zn [49]. These results indicate that fast sound is a general feature of liquids near

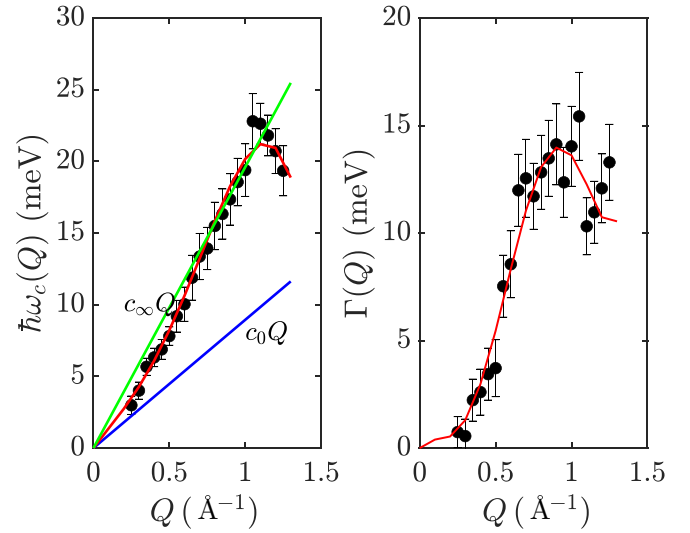


FIG. 4. Dispersion curve $\hbar\omega_c(Q)$ and damping parameter $\Gamma(Q)$ obtained from the DHO fits for D_2S (symbols). The straight lines represent the adiabatic sound dispersion $\hbar\omega = c_0Q$ (blue line) and the fast-sound branch $\hbar\omega = c_\infty Q$ (green line). The red line is a guide to the eye.

their melting points, although its magnitude depends strongly on intermolecular interactions. Molecular liquids without extended hydrogen-bond networks typically show $c_\infty/c_0 \approx 1.5$, whereas hydrogen-bonded liquids reach values around ~ 2 .

It is noteworthy that a collective mode with a short lifetime (large damping) is observed throughout the explored momentum range. Although the damping is larger than in water and other HB liquids, the ratio $\eta(Q) = \Gamma(Q)/\omega_c(Q) \approx 1$ indicates that the mode is not overdamped. It is worth noting that the fitted energies $\hbar\omega_c(Q)$ are relatively high compared to the accessible dynamic range shown in Fig. 1. However, this does not compromise the fit: the maxima of the DHO occur at $\hbar\omega_c(Q)/\sqrt{1 + \eta(Q)}$, and since $\eta(Q) \approx 1$, the observed peaks lie below $\hbar\omega_c(Q)$ by about a factor $\sqrt{2}$.

Once the analogy with water is established for the main longitudinal excitation, a fundamental difference becomes evident: D_2S shows no sign of the weakly dispersive 5–7 meV mode characteristic of water. The complete absence of this feature, even in the high-resolution spectra, within the accessible (Q, ω) range, demonstrates that the secondary low-energy excitation observed in water is not a generic property of molecular liquids, but rather arises from the constraints imposed by hydrogen bonding. This agrees with the observation that the mode is more pronounced in neutron scattering [17] than in x-ray scattering [18]. Its absence in D_2S remains evident even in high-resolution data, where a weak or heavily damped signal would otherwise be detectable.

The ability to extract coherent excitations depends on the relative weight $f(Q)$ of the integrated coherent signal with respect to the total scattering. Figure 5 compares $f(Q)$ for D_2S (this work) with that of D_2O [17] and D_2SO_4 [61]. The comparison reveals that D_2S has a favorable coherent-to-total ratio in the low- Q region, facilitating the observation of the longitudinal mode. In contrast, the signal becomes almost

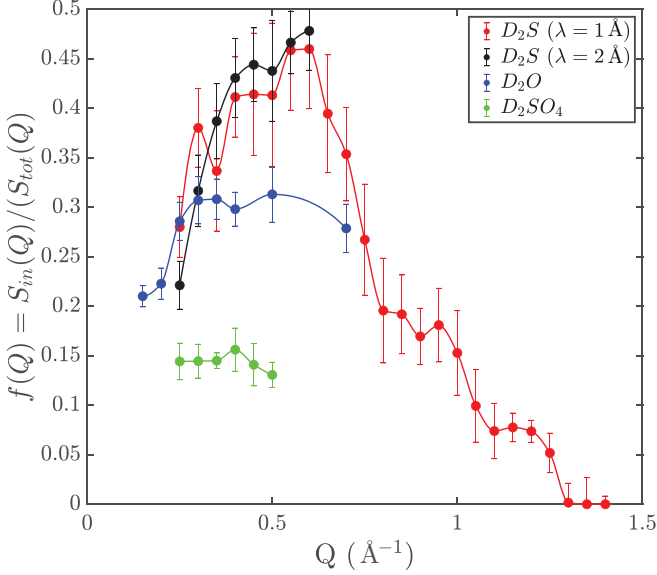


FIG. 5. Ratio between the energy-integrated inelastic (coherent) contribution and the total neutron signal for D_2S (this work), D_2O [17], and D_2SO_4 [61].

entirely incoherent at high Q , limiting the extraction of collective features.

B. Density of states in H_2S

The H_2S spectra shown in the right panels of Figs. 2 and 3 exhibit the typical features of an almost purely

incoherent neutron scatterer. The intensity is dominated by a broad, resolution-limited quasielastic peak associated with diffusive dynamics. No distinct inelastic side structures are observed, although a high-energy tail may indicate the onset of weak inelastic contributions above the explored range. This spectral behavior reflects the overwhelming dominance of proton incoherent scattering, which masks most of the collective features clearly visible in the deuterated sample.

The H_2S data were then exploited to extract the hydrogen-projected vibrational density of states $g_H(\omega)$. This quantity follows from the standard relation between $g_H(\omega)$ and the incoherent dynamic structure factor [82,83],

$$g_H(\omega) = \lim_{Q \rightarrow 0} e^{-2W(Q)} \frac{2M_p \hbar \omega}{\hbar^2 Q^2 [n(\omega) + 1]} S_{\text{inc}}(Q, \omega), \quad (3)$$

where M_p is the proton mass, $e^{-2W(Q)}$ is the Debye–Waller factor, and $n(\omega) = 1/(e^{\beta \hbar \omega} - 1)$ is the Bose population factor.

In practice, the extraction of $g_H(\omega)$ is constrained by the limited dynamic range of the experiment, which restricts the extrapolation at low Q . Furthermore, since the Debye–Waller factor is not straightforward to define in liquids, we adopt a simple approximate form $\exp(-0.004 Q^2)$, derived from the Q dependence of the quasielastic peak. Its effect is expected to be small, as this correction enters only at fourth order in Q .

The hydrogen-projected density of states obtained at both wavelengths [$\lambda = 1 \text{ \AA}$ and $2 \text{ \AA}$] is shown in Figs. 6(a) and 6(b). These spectra represent the hydrogen dynamics in liquid H_2S up to about 25 meV ($\sim 6 \text{ THz}$) and are compared [see panel (a)] with the corresponding quantities of liquid D_2S , liquid water [61], and solid H_2S [72]. Unlike the other

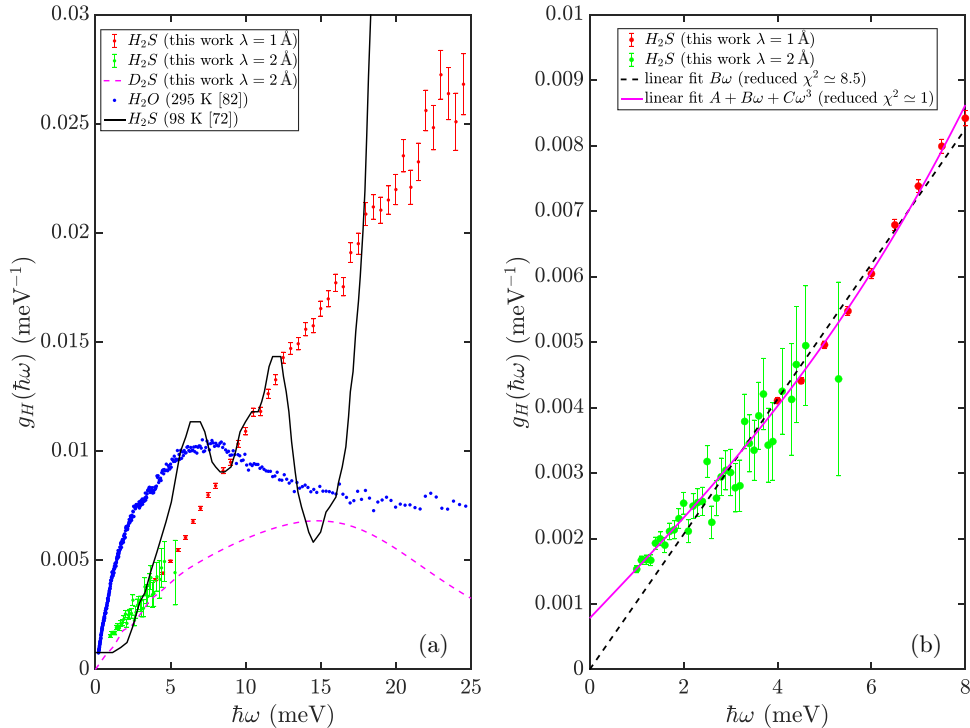


FIG. 6. (a) Hydrogen-projected DoS $g_H(\omega)$ for H_2S at $\lambda = 1 \text{ \AA}$ (red) and $\lambda = 2 \text{ \AA}$ (green), compared with liquid water at 295 K (blue) and solid H_2S at 98 K [72] (black). The dashed magenta line corresponds to the inelastic contribution extracted from D_2S coherent scattering. (b) Comparison between the experimental $g_H(\omega)$ and polynomial fits (linear and cubic). Reduced χ^2 values are reported in the legend.

systems, the DoS of liquid H₂S is largely featureless within this range, reflecting the predominance of whole-molecule motions. Isotopic effects are minimal; accordingly, the same low-energy behavior is expected in D₂S, consistent with the dynamic structure factor shown in Fig. 2.

The DoS increases smoothly with energy with an approximately linear trend. This behavior is characteristic of liquids with freely rotating molecules and stands in sharp contrast to the mode accumulation observed in hydrogen-bonded liquids.

In H₂O, by comparison, a strong accumulation of vibrational modes appears between 3 and 10 meV, mirroring the secondary mode detected in coherent scattering. No analogous feature is present in H₂S. The $g_H(\omega)$ extracted from the D₂S coherent spectra exhibits a broad bump at intermediate energies, consistent with the dispersive longitudinal mode identified in Fig. 4.

A comparison with the vibrational DoS of solid H₂S [72] provides additional insight. At 120 to 187 K, the solid adopts a face-centred-cubic, orientationally disordered structure similar to the liquid. Below about 110 K, molecular orientations freeze, and sharp features emerge in the DoS. The 98 K spectrum displayed in Fig. 6(a) shows a series of peaks absent in the liquid, reflecting the formation of distinct lattice modes.

This comparison confirms that the liquid lacks the structural constraints responsible for the 5–10 meV mode accumulation observed in water, reinforcing the central role of hydrogen bonding in shaping the low-energy vibrational spectrum of HB liquids.

Figure 6(b) reports the low- ω behavior of the DoS of liquid H₂S. Recalling the relation between the $\omega \rightarrow 0$ limit of $g(\omega)$ and the diffusion constant D [84],

$$g_H(0) = \lim_{\omega \rightarrow 0} g_H(\hbar\omega) = \frac{2}{\pi\hbar} \frac{DM_p}{k_B T}, \quad (4)$$

which holds for water [59], we note that the expected linear behavior $g(\omega) \propto \omega$ proposed in [85,86] is modified in the low-energy region because of the self-diffusion contribution. To accurately examine the low- ω behavior of $g(\omega)$, we performed low-energy fits of the data collected at $\lambda = 2 \text{ \AA}$ together with those measured at $\lambda = 1 \text{ \AA}$. These fits show that the spectra are not compatible to $g(0) = 0$, highlighting a finite low-frequency contribution. Fits constrained to $g(0) = 0$ give a poor reduced $\chi^2 \approx 8$. A cubic polynomial gives $g(0) = (0.00085 \pm 0.00013) \text{ meV}^{-1}$, compared to the estimate $(0.00211 \pm 0.00013) \text{ meV}^{-1}$ from Eq. (4). Although the two values differ, the contribution of self-diffusion must be considered. After performing several tests by varying the fit range and the polynomial order, we found that a cubic form $g(\omega) = A + B\omega + C\omega^3$, with a fixed value $g(0) = A = (0.00085 \pm 0.00013) \text{ meV}^{-1}$, provides the best fit, with $\chi^2 \simeq 1$. The estimate (0.00211 ± 0.00013) obtained from Eq. (4) is larger; however, the low-energy scattering data are affected by the experimental resolution, which broadens the quasielastic peak, an effect that becomes increasingly significant in the low- (Q, ω) region. A quantitative estimate of the diffusion coefficient using Eq. (4) is, in principle, possible; however, in the present case, it would be strongly affected by the limited energy resolution and is therefore not pursued.

This behavior is consistent with recent measurements on several liquids, including water, sulfuric acid, and Fomblin

[87]. Although the most recent study does not strictly follow the standard relationship between the velocity auto correlation function and the energy spectrum, the reported data, together with many other results from independent sources, tend to support an approximately linear dependence of the density of states on energy in the low-energy region. This behavior appears to be present regardless of the complexity of the liquid. This specific effect, observed through the formally exact extrapolation $Q \rightarrow 0$ of the incoherent dynamic structure factor, may be related to the fact that atomic dynamics in liquids is influenced by self-diffusion—an effect that may be progressively suppressed near the triple point, as is the case in the present experiment. These works support an approximately linear low-energy DoS in liquids, regardless of structural complexity, although the influence of self-diffusion becomes more subtle near the triple point—relevant for the present conditions.

To further analyze the vibrational content, we consider the integrated number of modes per hydrogen atom,

$$N_H(\omega) = \int_0^\omega g_H(\omega') d\omega', \quad (5)$$

which approaches three in the high-energy limit.

Up to 20 meV, liquid H₂S accumulates only about 5% of the total vibrational modes, consistent with the fact that most modes arise from high-energy intramolecular bending and stretching vibrations. At low energies, hydrogen motion is dominated by the rototranslational dynamics of the entire molecule, giving an expected mode fraction close to $M_H/(2M_H + M_S) \approx 0.0296$, in good agreement with experiments.

The low-energy vibrational content of liquid H₂S at 205 K is comparable to that of the orientationally ordered solid at 98 K, though the latter exhibits several sharp peaks. Heavy water displays a similar number of low-energy modes to D₂S up to about 10 meV; above this threshold, the lighter mass of H₂S shifts its modes to higher energies. The integrated contribution of the D₂S collective mode accounts for roughly 40% of the low-energy mode count observed in water.

The ratio $R(\hbar\omega) = N_H(\omega)/N_H^w(\omega)$, shown in Fig. 7, is close to unity for both liquid and solid H₂S, indicating similar low-energy vibrational content. Here, $N_H^w(\omega)$ denotes the corresponding quantity evaluated from the hydrogen-projected density of states of liquid water (H₂O or D₂O), enabling a direct comparison between the two systems. This near-unity ratio highlights that the absence of hydrogen bonding eliminates the mode excess observed in water, bringing the low-energy content of H₂S close to that of a freely rotating molecular liquid.

For D₂S, $R(\hbar\omega) \approx 0.45 \pm 0.03$, consistent with the inverse molecular mass ratio $M_{H_2O}/M_{D_2S} = 0.555$, showing that its low-energy dynamics reflect collective motions of the entire heavy molecule.

IV. CONCLUSION

We have performed a neutron-scattering investigation of the coherent and incoherent THz dynamics of liquid hydrogen sulfide in order to clarify the role of hydrogen bonding in the dynamics of water.

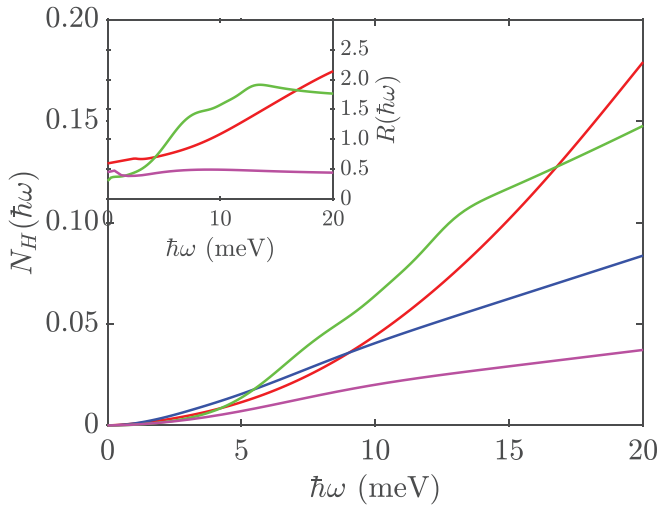


FIG. 7. Number of vibrational modes per hydrogen in liquid H_2S (red) as a function of energy. Green: solid H_2S at 98 K; blue: liquid water at 295 K [59]; magenta: D_2S inelastic contribution. Inset: ratio $R(\hbar\omega) = N_H(\omega)/N_H^w(\omega)$ for H_2S (liquid and solid) and for D_2S .

A key result from the coherent dynamics of D_2S is that the absence of hydrogen bonding does not suppress the fast-sound phenomenon: a longitudinal mode with velocity $c_\infty \gg c_0$ is clearly observed, similar to the behavior found in water and in many hydrogen-bonded liquids. This demonstrates that fast sound is not a unique fingerprint of hydrogen-bonded liquids, but rather a more universal dynamical feature of molecular fluids in the THz regime. The inelastic signal is visible across the entire measured Q range, and the modeling closely follows that established for heavy water.

The second major finding is the complete absence, in D_2S , of the secondary weakly dispersive 6–7 meV mode that is a hallmark of water and other hydrogen-bonded systems. The absence of this excitation in a structurally similar but nonhydrogen-bonded liquid shows that it arises from hydrogen-bond-stabilized local structures, where restricted molecular reorientations generate solid-like vibrational behavior.

The present results are consistent with the behavior expected for simple molecular liquids, where the high-frequency dynamics can be understood within a generalized

hydrodynamic or viscoelastic framework. In particular, similar features have been reported in simple liquids such as neon and argon, where the dynamic structure factor can be successfully described in terms of one extended heat mode and two damped sound modes, without invoking additional propagation mechanisms [88]. A similar scenario has also been reported for other simple molecular liquids, such as CO_2 , where inelastic neutron scattering and molecular dynamics simulations consistently show that collective excitations can be accurately described within a viscoelastic framework, with a clear positive dispersion of the sound velocity [89]. Within this picture, the apparent increase of the sound velocity at finite wave vector has a viscoelastic origin, arising naturally from the crossover between hydrodynamic and solid-like response. In this respect, the behavior observed in H_2S is consistent with earlier studies by Balucani *et al.* [42], where it was shown that, despite the molecular similarity with water, H_2S behaves more like a simple liquid such as argon. In particular, the absence of a hydrogen-bond network results in a substantially weaker anomalous dispersion, in agreement with the present findings.

Finally, the incoherent dynamics of H_2S also highlight the dominant role of hydrogen bonding in shaping the low-energy density of states in water. Whereas H_2O exhibits a clear accumulation of modes at 6–7 meV, the DoS of H_2S is smooth and featureless, consistent with the absence of hydrogen bonding and with more freely rotating molecules. Taken together, the two complementary isotopic measurements show that hydrogen bonding governs the fine structure of the low-energy spectrum, while the fast-sound behavior arises from more general features of liquid structure and dynamics.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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