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The Doppler effect in NMR spectroscopy

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Abstract

An NMR sample may be subject to motions, such as those due to sample spinning or to liquid flow. Is the spectrum of such a sample affected by the Doppler effect? The question arises because, instrumental dimensions being much shorter than the wavelength, it is the near-field of the precessing magnetic moment which couples to the receiver coil, rather than the radiated far-field. We expand the near-field into plane propagating waves. For each such wave there is another one with the same amplitude, propagating in the opposite direction. The Doppler shifts are therefore equal and opposite. In the model case of a small fluid sample moving with constant velocity, this leads to a distribution of Doppler shifts which is symmetrical with respect to the unshifted frequency: there is no net spectral shift. We examine the possibility of observing the Doppler distribution in this case. We also consider the case of thermal motion of a gas. We draw attention to the resolved Doppler splitting of molecular rotational transitions in a supersonic burst as observed in a microwave resonator. We also mention briefly the Doppler effect in molecular beam spectroscopy.

Keywords: Doppler shift; Doppler effect; Molecular beams; Molecular rotational spectroscopy; NMR

1. Introduction

The Doppler effect is a general spectroscopic property of systems in motion. It consists in a shift δf of the frequency f of the traveling wave recorded by a receiver in translational motion relative to the source. In a common formulation, the classical Doppler shift δf is given by:

$$\delta f/f = v_{//}/c, \tag{1}$$

where $v_{//}$ is the projection of the velocity v of the source on the direction from the source to the receiver and c is the velocity of light. Relativistic corrections to this result are of second order in v/c.

Doppler radar measures car speed on highways, and differentiates between mobile planes and static background in airports. The tracking of a space probe makes use of the Doppler shift of the probe's radio transmitter. In Mossbauer spectroscopy the Doppler shift is used to sweep the nuclear γ-ray transition in the moving emitter or absorber. The Doppler effect shifts the light of an approaching star to the blue, of a receding star to the red. It broadens the spectral lines of atoms or molecules in a gas due to their random thermal motion.

In all of these cases, the distance between source and receiver is many wavelengths, and the receiver is detecting a quasi-planar traveling wave radiated by the source. The first-order Doppler shift is easily derived by counting the oscillations of the electromagnetic field at the receiver in a given time interval. For instance, this number is greater when the source is approaching the receiver, because, as the source gets closer, the field has a shorter distance to travel, so the delay between emission and reception is constantly diminishing, hence the increased frequency.

NMR spectroscopy deals with the radio-frequency magnetic field generated by nuclei precessing in a static field $\mathbf{B}_0$, and, reciprocally [1–3], with the coupling of nuclei to an external radio-frequency field $\mathbf{b}_1$. Often the nuclei are moving, as in a spinning sample, liquid or solid, or in a flowing biological fluid. In the case of a spatial variation of the instrumental static or radio-frequency (rf) magnetic fields, sample motions may affect NMR spectroscopy. For instance, diffusion is measured by the loss of transverse magnetization in a field gradient. Inversely, sample spinning averages out the static

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field inhomogeneity, thus narrowing the resonance line. When the direction of the rf field is radial, sample rotation shifts the NMR frequency [2,4]; with a different rf field geometry, the same effect may be used to measure flow [5]. A different case is that of solid-state NMR, where sample rotation modulates internal interactions such as the nuclear dipole–dipole interaction, generating important spectral modifications [6].

None of these effects of sample motion is related to the Doppler effect. But is a Doppler effect also present, and if so, would it be detectable?

For instance, consider a liquid sample in the typical 5 mm o.d. tube of a 500 MHz high-resolution NMR spectrometer. If the tube spins at 50 Hz, v/c is about 2×10^{-9} at the periphery, so that δf is 1 Hz. In this case, one would not expect a net shift, because the speed vector of a given element of the sample rotates with time and is zero on the average. Furthermore, the coherent modulation of the presumed Doppler shift would lead only to spinning side-bands at the rotation frequency and multiples. In the case of solid-state NMR, the rotation frequency and the velocity are about 100 times larger. But the effect of the Doppler shift would again be limited to contributions to the rotation side bands which could be difficult to distinguish from others.

There are few references to the Doppler effect in the NMR literature, probably because the contemplated effect would be rather small, although not out of reach of present techniques, as indicated by the computation above. An NMR Doppler shift was considered briefly for a flowing liquid in a physiological context [5]. We found no mention of a Doppler effect related to sample spinning, even though it might in principle perturb this line-narrowing procedure. In NMR of gases, the Doppler effect due to thermal motion would produce no shift, since the average velocity is zero, and this result is therefore mute on the question of a Doppler shift in the case of uniform velocity. A Doppler shift related to the net velocity of atomic or molecular beams could be important for the high precision clocks based on nuclear, atomic or molecular transitions. We shall return to molecular beams below.

For a simple case, one could set up a flow of water at constant speed parallel to the static field within a capillary tube (Fig. 1). Slice-selective excitation would create transverse magnetization in a small lump of water slightly downstream of the rf coil, and one would then record the free precession.

Even in this simplest NMR situation, one wonders whether or not a Doppler shift occurs. On one hand, one might say that the Doppler shift is a consequence of special relativity and is therefore universal. On the other hand, one notes that the receiver couples mainly to the near-field of the sample source rather than to a radiated progressive wave. Indeed, sensitive detection of the free precession occurs only as long as the sample is within a

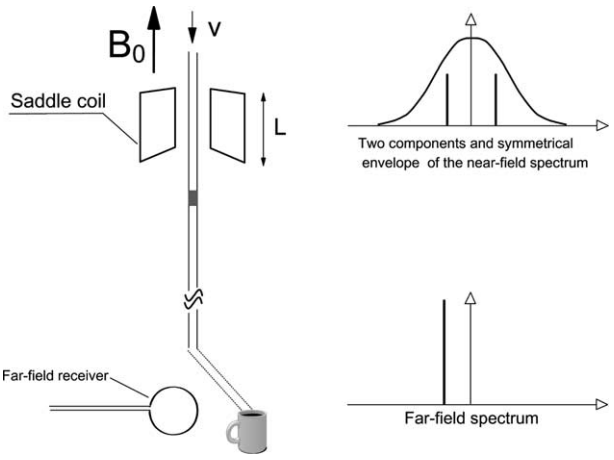

Fig. 1. Principle of the measurement of the NMR spectrum of a small lump of water moving at constant speed. Water flows through the vertical tube. At time zero, an rf pulse in the saddle coil flips the proton magnetization by 90° in a small region (in gray), selected by a strong field gradient pulse. The free precession of the magnetic moment of the protons in the lump is recorded by the same coil. In both cases, coupling is by the near-field. A distant antenna would receive the radiated far-field of the free precession, which is much stronger than the non-radiative near-field at the position of the antenna, but much weaker than the near-field at the position of the coil. The "down-field" shift in the far-field spectrum corresponds to a positive Doppler shift.

range R comparable to the size of the receiver coil, typically 1 cm. Since this is much less than the wavelength (60 cm), the near-field dominates. Thus, one might argue against a Doppler shift, since this shift is a property of progressive waves.

One feature of the problem is the limited time available for the measurement, namely the time T during which the lump remains within range of the receiver, $T \approx R/v$. This will lead to an instrumental broadening W:

$$W = 1/(2\pi T) \approx v/(2\pi R). \tag{2}$$

Combining Eqs. (1) and (2), and introducing the wavelength $\lambda = f/c$, we get:

$$\delta f/W \approx 2\pi R/\lambda. \tag{3}$$

Since the near-field situation requires that R be comparable to or smaller than λ, the shift would not be much larger than the instrumental broadening, at best. This would complicate detection of the shift, but it would not make it impossible. So the question remains: is there a Doppler shift of the signal of the lump moving at constant speed? In the present note, we show that in the case of near-field coupling, the answer is not yes or no, but may amusingly be stated as: both yes and no.

2. Theory

We ignore any feature specific of the nuclear spins. We consider the classical precessing magnetic dipole resulting from the precessing nuclear moments in the

small water lump. Furthermore, a precessing magnetic dipole may be built up from two oscillating dipoles making an angle of 90° and oscillating in quadrature. We therefore consider only an oscillating dipole.

2.1. The resonant frequency of a moving sample

We note first of all that the static magnetic field $\mathbf{B}_0$ is the same, to first order in v/c, in the reference frames linked to the laboratory and to the moving sample: in a Lorentz transformation, the magnetic field component parallel to the velocity is unaltered; and in the absence of an electric field, the change in the perpendicular component is second order in v/c [7]. Therefore, the Larmor frequency $f = \gamma B_0/(2\pi)$ in the reference frame of the sample is unchanged to first order in v/c.

2.2. The field of an oscillating magnetic dipole

As is well known, the magnetic and electrical fields (**b**, **e**) created at a given point R by a dipole located at point S and oscillating at frequency f may be separated into two components. One component, the far-field, is a radiated wave. As such, its phase is position-dependent, and its amplitude, as one moves away from the dipole in a given direction, varies as the inverse first power of the distance from the dipole. The spatial distribution of the second component, the near-field, is quite similar to the magnetostatic field of a constant dipole. Its amplitude varies as the inverse distance cubed, and its phase is independent of position. Of course, both components oscillate at frequency f.

It is useful to give a complementary description of the dipole fields. We expand them into plane propagating waves, each of which separately obeys Maxwell's equations. Since the frequency is the same for them all, so is the modulus of the wave vector, $k = \omega/c$. At a point P distant from the dipole, the magnetic field **b** is well represented by a quasi-planar wave whose phase is $\exp(\mathrm{i}(\mathbf{kr} - \omega t))$, with the wave vector **k** oriented along, or nearly along, the SP direction (unit vector **u**). The magnetic field is perpendicular to **u**, and for a given direction **u,** its amplitude is inversely proportional to the distance between S and P. The electric field of the wave is perpendicular to **u** and **b**. This wave propagates away from the source.

At a point P' close to the dipole, the near-field dominates. In the expansion into plane waves, the condition of a position-independent phase requires that waves with opposite propagating vectors, **k** and **−k,** have equal amplitudes (see Appendix A). Hence each couple of waves, **k** and **−k**, corresponds to a standing wave. The occurrence of waves with opposite wave vectors implies that at each point, for each wave which propagates away from the source there is another one propagating towards it.

Given the geometry of the dipole fields, it might seem more appropriate to expand the fields into cylindrical waves rather than into plane waves. Still, both expansions are possible since each type of waves forms a complete set. The compelling reason for choosing plane waves in the present case is that such waves transform simply in a change of the Galilean reference frame: they remain plane waves. The frequency is Doppler-shifted and the direction of the wave is subject to an aberration which is first order in v/c.

2.3. The fields of a moving dipole

How do the fields of the dipole transform when viewed from a reference frame (the laboratory) which is in motion with respect to the dipole? To answer the question, we simply transform each of the plane wave components of the dipole field. Each one is subject to a Doppler effect which is given by Eq. (1), with however one *caveat*: this equation applies to the far-field situation, where the direction from source to receiver is the same as the direction of the wave vector of the wave received from the source. But, in the general case, the wave vectors of the planar components of the dipolar field point in different directions, including, as we saw above, some which point towards the source. One must therefore consider whether one should interpret $v_{//}$ in Eq. (1) as the projection of the source velocity along the source-to-receiver direction or along the direction of the wave vector. A plane wave is characterized by its 4-(wave vector) which combines **k** with $\mathrm{i}\omega/c$ as the fourth component. For the Lorentz-transformed plane wave, this component has the value $\mathrm{i}(\omega/c + \mathbf{v}\cdot\mathbf{k}/c^2)$, to first order [8]. Since $k = \omega/c$, one obtains the general expression for the Doppler shift:

$$\delta f/f = (v/c)\cos\theta, \tag{4}$$

where θ is the angle between the wave vector and the velocity of the source. Thus, in Eq. (1), $v_{//}$ should be considered as the projection of the velocity of the source along the wave vector.

In the case of the far-field, Eq. (1) follows from Eq. (4). In the case of the near-field, which is the one which applies to the NMR situation, each plane wave component is shifted according to the value of cos θ. For each component with a positive shift, there is another with the same amplitude and with the opposite shift. Further discussion of the far-field and near-field may be found in Appendix B.

2.4. The signal induced by the dipolar field in the receiver coil

The signal is the sum of the electromotive forces induced in the coil by the plane wave components. The sensitivity of the coil depends on the orientation of the

magnetic field of the wave with respect to the coil. Furthermore, the sensitivity is not necessarily the same for two waves propagating in opposite directions, a property put to good use for instance in the omni-present Yagi television antennas. However, equal sensitivity for opposite wave vectors does obtain in the case of a passive structure which is small compared to the wavelength.

The spectrum of the NMR signal induced in the coil by the near-field of a lump moving with uniform velocity therefore consists not of one unshifted line, nor of one Doppler-shifted line. Instead, it consists of two distributions, one which spans the range between the unshifted Larmor frequency f and the Doppler-shifted frequency $f(1+v/c)$ (Eq. (4)), and a second one which is its mirror image, spanning the frequency range between f and $f(1-v/c)$. The spectrum is furthermore broadened according to Eq. (3). Note that the ratio of the Doppler shift to broadening is independent of the velocity v. In conclusion the spectrum is affected by the Doppler effect but the center of the frequency distribution is unshifted.

2.5. Observability of the near-field NMR Doppler spectrum

If there had been a net Doppler shift in the case of the sample with uniform velocity, one could probably have arranged to measure it, even in the presence of the instrumental broadening W and despite the small value of the Doppler shift, ca. 1 Hz as computed above. The detection of the symmetrical Doppler distribution seems more difficult. However, since a net Doppler shift is not to be found, there is no reason anymore to consider only uniform motion. We may just as well consider the Doppler effect of non-uniform motions, in which the sample may be maintained permanently within receiver range, so that the instrumental broadening (given by Eq. (2) in the case of uniform motion) vanishes. The case of a rotating sample was mentioned above. It presents the difficulty that the Doppler shift brings in no new frequency but only contributes to the spinning side bands, and it may be difficult to distinguish this contribution from others.

Another case is that of a gaseous sample. The thermal velocity is high, but frequent interatomic collisions slow down the (diffusive) escape of an atom out of receiver range. In conditions of fast exchange, the collisions induce line-narrowing. The linewidth Δf is given by [9]:

$$2\pi\Delta f \approx (2\pi\delta f)^2\tau \tag{5}$$

where τ is the correlation time for the direction of the velocity. By Eq. (1):

$$\Delta f/f \approx 2\pi f\tau(v/c)^2. \tag{6}$$

Consider for instance the nuclear resonance of ^{3}He. At room temperature, the average root mean square thermal velocity is about 1000 m/s, corresponding to a first order Doppler shift of 3.3×10^{-6}, so that δf is 1650 Hz for a Larmor frequency of 500 MHz. At room temperature and atmospheric pressure, τ is in the range of 0.1 ns, and $2\pi f\tau$ is about 0.3 so that $\Delta f/f$ is comparable to the second-order Doppler shift $(v/c)^2$. The width Δf is only 1.5 mHz. But the collision time is inversely proportional to pressure so that the Doppler width would rise to a detectable value of 1.5 Hz at a pressure of one milli-atmosphere, a pressure still large enough to provide acceptable sensitivity.

The inverse dependence of the Doppler width on pressure is helpful for another reason. It helps distinguish the Doppler contribution to the linewidth from the dipole–dipole contribution, the latter being directly proportional to pressure because the dipole–dipole interaction operates principally during collisions. Note that the dipolar contribution can be reduced by operating with a buffer gas such as ^{4}He.

3. An observation of Doppler splitting

A current method for the study of molecular rotational states provides a clear illustration of the spectroscopic effect of source motion in a near-field, non-radiative context [10]. A pulsed supersonic nozzle injects gas into a Fabry–Perot microwave cavity, tuned to the transition between two molecular rotational states, ca. 10,000 MHz. After the gas bursts out from the nozzle, the molecules travel in straight lines. The distribution of directions is broad, but the velocity distribution is narrow. In a study of the $J=0$–1 transition of the OCS molecule, the velocity, which is related to the average thermal velocity in the gas reservoir, was 400 m/s, which gives a Doppler shift of 13.33 kHz [11]. The transition was excited by the electrical field of a 6 μs microwave pulse, and the subsequent molecular response, lasting about 50 μs, was detected in the absence of any external field. The situation closely resembles that of excitation and detection of the free precession in a pulsed NMR (or EPR) measurement. The microwave cavity, like a NMR coil with its tuning capacitor, is a resonant circuit. And the coupling of the two molecular rotation states with electric fields is analogous to the coupling of a fictitious spin 1/2 with magnetic fields [12]. So again we ask: is the spectrum Doppler-shifted by the molecular motion? Is it split?

The answer, provided by a detailed theory and fully confirmed by experiment, is unambiguous [11]. The spectrum is split into two nearly identical lines, with maxima equally shifted from f_0, where f_0 is the frequency of the transition which is known independently. The two lines are well resolved, with a splitting of 36.3 kHz, which is 2.3 times the full width at half height. The line shape results from a Doppler shift distribution

which corresponds to the angular distribution of the molecular velocities. Such observations are more than academic since the method of the standing wave pulsed Fourier transform in combination with a pulsed supersonic nozzle is among the best current methods for the spectroscopy of short-lived molecular species [10].

The interested reader is referred to the original publications for further information. Here we mention briefly three features of supersonic pulse rotational spectroscopy which contribute to a resolved Doppler splitting. First, the large molecular velocity, combined with the high frequency of the rotational transition, provides for the large Doppler shift. Together with the long relaxation times of the transition, this insures that intrinsic broadening is small compared to the linewidth related to the Doppler effect itself. Second, the cavity detector provides stationary fields over lengths comparable to or larger than the wavelength, so that the relative instrumental broadening W/δ (Eq. (3)) may be smaller than in an rf coil. Third, the cavity mode is more directional than the rf coil: it is a standing wave consisting of two quasi-planar waves propagating in opposite directions, so that the expansion into plane waves involves mostly components whose wave vector is close to one of a couple of wave-vectors $\pm\mathbf{k}$, in contrast to the broader distribution of $\mathbf{k}$ orientations in the case of the rf coil. The distribution of Doppler shifts on each side of the unperturbed frequency is therefore narrower.

4. Molecular beams

In molecular beam spectroscopy, a beam of particles (molecules, atoms, ions...) with fairly uniform velocity, typically in the range of 100 m/s, moves past one or more regions where an rf field induces transitions between states of the particle. Among many applications of this method, one may mention the measurement of magnetic moments, and the cesium atomic clock. Such applications would be sensitive to a Doppler shift and to the Doppler broadening resulting from a velocity distribution. In the case of atomic clocks, even the second order Doppler shift must be taken into account [13,14]. The discussions of the Doppler effect in the early literature are often brief. In "Molecular Beams" [15], it is pointed out in Section V.6 which treats of resonance distortions, that a (radio-frequency) standing wave corresponds to a superposition of two running waves and that the Doppler effect will broaden but not shift the resonance if the amplitudes of the two waves are equal. This section carries a reference to a report on Doppler splitting in an atomic beam passing through a waveguide with a standing wave pattern [16]. A related observation is that a sufficient condition for the absence of a net Doppler shift is that the phase of the field be constant. One way to satisfy this condition is to use a field region which is small compared to the wavelength. This is possible without drawbacks in the separated oscillatory field method.

In the same section, another distortion is mentioned, the so-called Millman effect. This is a resonance asymmetry due to a frequency shift which occurs if the direction of the rf field changes with position [17]. This is closely related to the frequency shift under sample spinning in a radial rf field [2]. In particular, the dependence on the sign of the nuclear moment was noted in both reports. Regretfully, the later report did not quote the earlier one. The Millman effect is independent of the resonance frequency, in contrast to the Doppler effect.

5. Conclusion

Perhaps surprisingly, a uniform motion of the NMR sample does not produce a net Doppler shift of the resonance detected in the usual near-field situation. It does produce a symmetrical shift distribution, but the corresponding line broadening would be difficult to observe. On the other hand, it may be possible to detect the NMR Doppler broadening due to thermal motion in a gas. The validity of the present analysis is supported by a related phenomenon, the Doppler splitting of molecular rotational transitions as observed in a microwave resonator.

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Appendix A. Plane wave expansion of a monochromatic field with position-independent phase

The monochromatic rf magnetic field $\mathbf{B}(\mathbf{r})$ is given by:

$$\mathbf{B}(\mathbf{r}) = \mathbf{b}(\mathbf{r})\exp(2\pi \mathrm{i} f t).$$

For a position-independent phase, the phase of $\mathbf{b}$ must be independent of $\mathbf{r}$. We may then choose $\mathbf{b}$ real. We expand $\mathbf{b}$ into linearly polarized plane waves, which form a complete set:

$$\mathbf{b}(\mathbf{r}) = \sum \mathbf{a}(\mathbf{k})\exp(\mathrm{i}\mathbf{k}\mathbf{r})\mathbf{dk}.$$

Since we are dealing with electromagnetic waves in vacuum, the waves are transverse, so that $\mathbf{a}(\mathbf{k})$ is perpendicular to $\mathbf{k}$, and the summation $\sum$ is over two perpendicular polarization states. Since $\mathbf{b}(\mathbf{r})$ is real, it is equal to its complex conjugate. This relation is valid for

all **r,** and hence for each **k** and each polarized component. Hence

$$\mathbf{a}(\mathbf{k}) = \mathbf{a}^*(-\mathbf{k}).$$

This shows in particular that for each plane wave component propagating in the **k** direction, there is one in the opposite direction, and their amplitudes are equal. Each such couple will generate two peaks of equal amplitudes, with opposite Doppler shifts. As a result, there is a symmetrical distribution of Doppler shifts: the Doppler effect gives rise to a broadening with zero average shift.

Appendix B. Far-field and near-field

The Doppler shift of the wave emitted by an oscillating dipole in motion is closely related to the retardation of the fields. This is emphasized in Feynman's text on the fields of a moving charge which is the basis for the present appendix. Since a point magnetic dipole is equivalent to a minute current loop, and therefore to moving charges, the principal features of the fields are common to dipole and charge. They are illustrated by the transformation properties of fields due to a moving charge q.

Starting from the Lienard–Wiechert formulae for the potentials, the fields are obtained as functions of the coordinates of the charge at the retarded position and time. Feynman derived the following expressions [18]:

$$\mathbf{E} = q/(4\pi\varepsilon_0)[\mathbf{e}/r^2 + (r/c)\mathrm{d}(\mathbf{e}/r^2)/\mathrm{d}t + (1/c^2)\mathrm{d}^2(\mathbf{e})/\mathrm{d}t^2],$$

$$\mathbf{B} = \mathbf{e} \times \mathbf{E},$$

where **B** and **E** are the fields at a position F and at time t_0. The symbol $\times$ indicates the vector product. The units are SI, and $4\pi\varepsilon_0 c^2 = 10^7$.

The fields are due to the source which, at time t_1, is located at point S_1 such that the distance r between S_1 and F is equal to $(t_0 - t_1)/c$. The unit vector **e** points from S_1 to F. The fields are "retarded" in the sense that the source at S_1 (time t_1) contributes to the field at the later time $t_0 = t_1 + r/c$. (One may note that the relation of t_1 to t_0 is implicit, and is defined by the trajectory of the source [19]. This is not a problem. Furthermore, to first order in v/c, t_1 is equal to the explicit value $t_1' = t_0 - r_0/c$ where r_0 corresponds to the position S_0 of the source at time t_0.)

The electric field is the sum of three terms. The third term, which varies as the inverse of the distance from the field-point to the source, gives rise to radiation, which is Doppler-shifted as a direct consequence of retardation. The first term is the (retarded) Coulomb field, which decreases as the inverse square of the distance and which one is tempted to consider as the main source of the near-field. Since it is retarded, it is also Doppler-shifted. But the second term also contributes to the near-field. For short distances, its main effect is to nearly cancel the retardation of the first, so that the near-field is nearly devoid of retardation! For instance, in the case of the field of an element of current, the sum of the first and second terms is much closer to the electrostatic law of Biot and Savart than to the same law modified by retardation. In Feynman's words: [This] "gives fields very much like the instantaneous theory—much closer than the instantaneous theory with a delay; the first-order effects of the delay are taken out by the second term. The static formulas are very accurate, much more accurate than you might think." In contrast to the far-field, the near-field from a moving source is not retarded.

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