

Raman spectrographic system for quantitative analysis of isotopic hydrogen mixtures for muon catalysis experiments

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We show that Raman spectral lines from H_2 , D_2 , T_2 , HD, HT and DT are readily resolved, permitting an effective means to analyze isotopic hydrogen mixtures used in muon-catalyzed fusion experiments. We propose a Raman spectrographic system to allow for real-time analysis of targets involving all three isotopes of hydrogen.

1. Introduction

Early on in our research, we found that resonant muo-molecular ion formation depends on the isotopic hydrogen molecular species present in the target in muon catalysis experiments [1,2]. Indeed, disentangling the rates of various muonic reactions during muon catalysis requires quantitative information regarding the concentrations of H_2 , D_2 , T_2 , HD, HT and DT.

We approached this problem during experiments at LAMPF a decade ago using separate reservoirs of D_2 and T_2 to permit on-site filling of the target at cryogenic temperatures. We found that D_2 plus T_2 mixtures gave much faster muo-molecular formation than did D_2 plus T_2 plus DT mixtures. In this way, we were able to achieve up to 150 d-t fusions per muon (average) [3]. We also demonstrated that $dt\mu$ formation in D_2 molecules proceeds much more rapidly, for temperatures up to 400 K, than does $dt\mu$ formation in DT molecules [2].

Muon-catalyzed fusion experiments were performed at LAMPF in the same time period using triple mixtures involving H, D and T. But without better means of determining the concentrations of molecular species present in real-time, we were unable to extract reliable information regarding $dt\mu$ -molecular formation rates in HT (for example). Therefore, we began to consider other means to establish these concentrations. We came to the conclusion that Raman spectroscopy offers much better means of determining isotopic concentrations than does mass spectrometry, gas chromatography, or infrared spectrometry for reasons which will shortly become clear.

Raman scattering peaks of the hydrogen isotopic molecular species are displayed

in fig. 1, generated with data extracted from refs. [4–6]. The figure demonstrates that Raman scattering provides several well-separated lines which permit quantitative identification of the species present.

Based on these data, we propose to use the most intense line for each of the species, with the exception of the HD(1) line which is within 1 cm^{-1} of the DT(3) line and therefore cannot be used to establish the HD concentration. For the determination of the concentration of HD, we intend to use the HD(0) line which is well-separated from other lines, and has an intensity 87.1% that of the HD(1) line [6]. The concentration of each hydrogen species can be calculated using the fact that the intensity of each line is proportional to the number of molecules of each in the mixture of H_2 , D_2 , T_2 , HD, HT and DT (or a subset thereof).

On the other hand, the presence of double transitions in infrared spectroscopy adds greatly to the complexity of the collision-induced spectrum which must be disentangled [4]. Furthermore, the Raman method is much faster than either mass spectrometry or gas chromatography, allowing for target-composition analysis in approximately 100 seconds [6], and without consuming any sample gas.

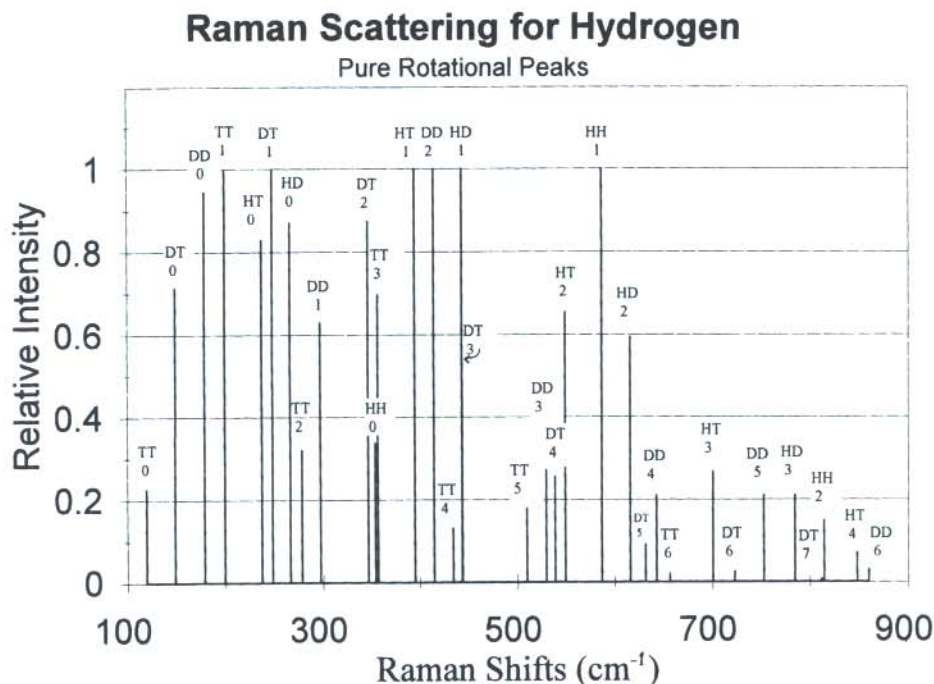


Fig. 1. Juxtaposition of Raman shift peaks for H_2 , D_2 , T_2 , HD, HT and DT for pure rotational states, i.e., for $\Delta v = 0$. The number below each hydrogen molecular species specifies the lower J value involved in the transition. The most intense line for each molecular species is normalized to one (1).

2. Proposed Raman system for muon-catalyzed fusion experiments

A schematic drawing of the proposed Raman analysis system is provided in fig. 2. Since the light scattering scales as λ^{-4} , it is desirable to use a short-wavelength light source. A standard 488 nm argon laser is well-suited to this purpose, although a mercury lamp with a band-pass filter may also serve.

The light enters the target through an optical window capable of withstanding high pressures in the target while transmitting the light efficiently. The choice of an optical window must take into account sealing the window into the target wall. Our studies indicate that a sapphire window would serve effectively, although optical fibers would also suffice. Scattered light contains radiation of the incident frequency ν (Rayleigh scattering) along with weaker radiation of frequency $\nu = \nu \pm \nu'$ (Raman scattering). The Rayleigh-scattered light is removed by an edge filter. The frequency difference ν' is characteristic of transitions in the scattering molecule and permits identification of the molecular species present (see fig. 1).

The Raman-scattered light emerges from the cell through the optical window and impinges on a set of six band-pass filters. Each filter is selected to transmit the Raman line for a particular molecular species, i.e., H_2 , D_2 , T_2 , HD, HT or DT. A photodetector (photodiode) behind each filter registers the intensity of the Raman line for each hydrogen specie. Alternatively, a monochromator may be used in conjunction with a photodiode array, as depicted in fig. 2. In either case, a single line is selected for each species, as specified above. Then each signal is proportional to the number of molecules present of each type, so that a quantitative analysis of the target composition is achieved in real-time. Since the molecules do not interact with

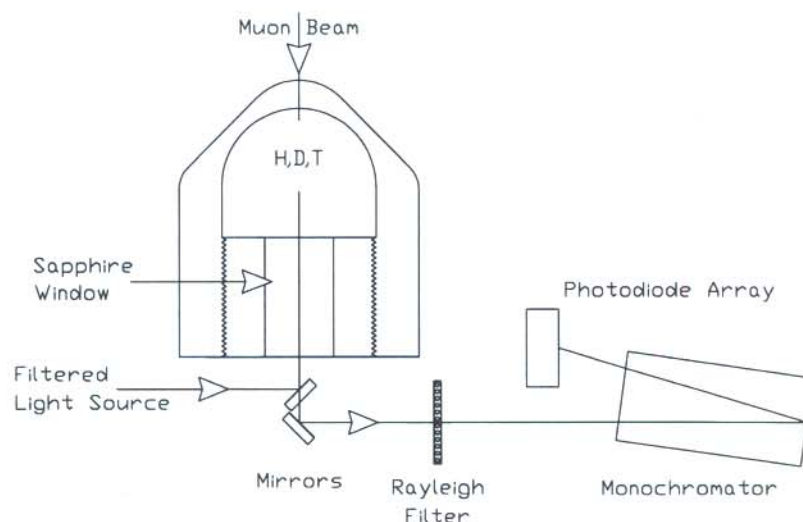


Fig. 2. Schematic of the proposed Raman spectrographic system for muon-catalyzed fusion experiments.

each other very much, at least at moderate pressures, line widths are narrow and easily resolved [4] (see fig. 1). As target gas density is increased, line broadening will increase approximately linearly, with $\Delta\nu \approx 2 \times 10^{-3} \text{ cm}^{-1}/\text{atm}$, approximately [7]. This line broadening is sufficiently small to allow good resolution of the Raman peaks for the different species up to pressures of approximately 5000 atm. Spectral information is computerized. The system may be calibrated using mixtures of known composition.

We propose, then, that the incident radiation be admitted directly into the target cell, with Raman-scattered light also collected through a target window, to permit real-time analysis of the molecular species present. In this way, it is not necessary to extract gas into a "Raman cell" for analysis, where the molecular composition may differ somewhat from that present in the target itself.

It is important to note that the Raman scattering increases linearly with the partial pressure of hydrogen, whereas the double transitions with infrared spectroscopy imply that the absorption intensity increases as the *square* of the partial pressure [4]. Thus, the Raman method succeeds with pressures as low as 0.05 kPa [5,6], where the infrared method could not be used. For solid hydrogens, 0.1–1 cm^3 is needed for either method [4].

In a test of a Raman spectrographic system somewhat similar to the one proposed here, Raman analysis required about 100 seconds whereas gas chromatography required 40 *minutes*; in both cases, hydrogen isotopes could be detected at a level of about 0.02% [6]. Clearly, Raman spectroscopy provides a preferred and highly satisfactory method for hydrogen-isotopic analysis for muon-catalyzed fusion experiments.

Using an edge filter to remove the Rayleigh-scattered light permits the spectrometer to function with a single monochromator (see fig. 2). By transmitting light directly into the target cell and gathering Raman-scattered light from it, real-time quantitative analysis of the isotopic hydrogen molecules present may be achieved. This system is specifically proposed for use in the muon catalysis experiments to be performed at Dubna, Russia.

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