Trace Detection of Metastable Helium Molecules in Superfluid Helium by Laser-Induced Fluorescence

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We describe an approach to detecting ionizing radiation that combines the special properties of superfluid helium with the sensitivity of quantum optics techniques. Ionization in liquid helium results in the copious production of metastable He₂ molecules, which can be detected by laser-induced fluorescence. Each molecule can be probed many times using a cycling transition, resulting in the detection of individual molecules with high signal to noise. This technique could be used to detect neutrinos, weakly interacting massive particles, and ultracold neutrons, and to image superfluid flow in liquid ⁴He.

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Renewed interest in superfluid helium as a particle detection medium has been spurred by proposals in low-background particle astrophysics (where superfluid helium has the advantage of extremely high radiopurity)[1, 2] and tests of the Standard Model using ultracold neutrons (where superfluid helium is used as an ultracold neutron production and storage medium[3, 4]). This paper introduces an evolution of these experiments by adding a component of direct triplet He₂ molecule detection through laser-induced fluorescence.

Experiments in the 1950s and 1960s showed that superfluid helium scintillates brightly in the extreme ultraviolet when exposed to ionizing radiation[5], and that large numbers of long-lived excitations are also created[6]. Detailed spectroscopy of electron-excited superfluid helium[7] later showed that both of these effects are due to the efficient production of He₂ excimer molecules, particularly in the He₂(1Σ⁺) and He₂(3Σ⁺) states which produce the scintillation and long-lived excitations respectively. It has been determined that more than 50% of the energy of an energetic electron in liquid helium is converted into chemical energy in the form of He₂ molecules, creating in total about 13000 He₂(3Σ⁺) and 19000 He₂(1Σ⁺) states per MeV[8]. Electronically excited helium atoms are unstable in liquid helium, rapidly reacting with ground-state helium atoms. For example, atoms in the 2S state, which have a radiative lifetime of 8000 seconds in vacuum[9], only survive 15 μs in the liquid due to non-radiative bonding with ground state atoms to form He₂(3Σ⁺) molecules[10].

While the lowest-energy singlet and triplet molecules both emit 80 nm photons when they radiatively decay[11], their radiative lifetimes are markedly different. The He₂(1Σ⁺) has a lifetime of about 1 ns[12], while the He₂(3Σ⁺) has a lifetime of 13 ± 2 s[13] in superfluid helium and forms a bubble of radius 5.3 Å[14]. The photon-emitting transition from He₂(3Σ⁺) to the dissociative ground state, He₂(XΣ⁺), is forbidden because it requires a spin flip. A theoretical value of 18 s for the lifetime of He₂(3Σ⁺) in the lowest vibrational state has been obtained by assuming the spin flip is driven by spin-orbit (SO) coupling and treating the Breit-Pauli spin-orbit Hamiltonian using first order perturbation theory[15]. The lifetimes of the lowest-energy triplet molecular states in neon, argon, krypton, and xenon are much shorter, measured to be 6.6 μs, 3.2 μs, 350 ns, and 50 ns respectively[16]. The large difference between the lifetimes of helium and the heavier noble gases is qualitatively due to the respective strengths of the SO coupling, which scales roughly as Z⁴. In addition, the lowest energy excited helium atom is in an S state, while the excited neon, argon, krypton, and xenon atoms are in P states. Therefore, the heavier noble gas molecules contain an intrinsic orbital angular momentum to contribute to the SO coupling that is lacking in helium molecules.

The metastable He₂(3Σ⁺) molecule can also decay through reactions with other helium molecules[10] and collisions with container walls. The movement of the molecule is limited by diffusive scattering with rotons, phonons and ³He impurities (see Figure 1). We model the diffusion constant by the following equation:

\[
\frac{1}{D} = e^{-\Delta/T} \frac{T^7}{\Gamma_r} + \frac{T^7}{\Gamma_p} + X \frac{T}{\Gamma_3}
\]

where X is the relative concentration of ³He in ⁴He, and \( \Delta \) is the roton energy gap (8.6 K). \( \Gamma_r, \Gamma_p, \) and \( \Gamma_3 \) represent measures of the scattering of the He₂(3Σ⁺) bubble with rotons, phonons, and ³He. \( \Gamma_r \) and \( \Gamma_3 \) have been determined experimentally[17], while \( \Gamma_p \) has been scaled from an experimental value for phonon-³He scattering by taking into account the greater effective mass of the He₂ bubble[14, 18]. By adjusting the temperature and ³He concentration, the displacement of He₂(3Σ⁺) over its radiative lifetime can be controlled, ranging from 0.7 cm to 40 m between 1 K and 100 mK.

Many detailed spectroscopic studies of triplet He₂ molecules in superfluid helium have been performed, and the energy levels and optical absorption frequencies are well established[2, 10, 19]. Figure 2 shows an energy level diagram of the lowest lying triplet states of the
the molecules to the $d^3\Sigma_u^+$ state, also resulting in the emission of 640 nm fluorescence. An advantage of this scheme is that only one laser is used. However, the blue excitation light is more likely than infrared light to induce spurious fluorescence in any associated optical elements, which could interfere with the detection of 640 nm $d^3\Sigma_u^+ \rightarrow b^3\Pi_g$ fluorescence.

When driving a cyclic transition in molecules, complications can arise from vibrational and rotational structure; if a large signal per molecule is needed, it would be undesirable to have molecules falling to a state in which they are not sensitive to the optical excitation frequency. Eltsov et al. have measured a vibrational relaxation time of $140 \pm 40$ ms for $a^3\Sigma_u^+$ molecules in superfluid helium, and they demonstrated that the rotational relaxation is much faster [19]. From calculation of Franck-Condon factors for the cycling transition shown in Figure 2, we have determined that only 1.2% of the molecules per cycle will fall radiatively to the first vibrational state rather than the zeroth vibrational state. If an experiment using this technique required the molecules to be cycled faster than the maximum effective vibrational relaxation rate of 600 s$^{-1}$, they can be repumped with a third laser operating at 1070 nm that would drive the molecules from $a(1)$ to $c(0)$ where they will decay to the $a(0)$ approximately 95% of the time. This repumping of the molecules into the ground vibrational state can be used to ensure that the molecules are sensitive to the optical frequency chosen for the cycling transition. In other experiments, it may be desirable to cause molecules created by one ionizing radiation event to be undetectable during a following event. In this case, the molecules can be driven into a different vibrational state where they will be blind to the pump lasers.

A detector using individual helium molecule fluorescence in a cycling transition would be far more sensitive to small energy depositions than a comparable scintil-
The differential cross-section for magnetic neutrino-electron elastic scattering is given by
\[
\frac{d\sigma}{dT}_{\text{MS}} = \frac{\pi \alpha_{\text{em}}^2 m_e^2}{m^2} \left[ 1 - \frac{T}{E_\nu} \right]
\]
where \(\alpha_{\text{em}}\) is the fine structure constant, \(m_e\) is the electron mass, \(T\) is the kinetic energy of the recoil electron and \(E_\nu\) is the neutrino energy \cite{22}. Because this scattering process is enhanced for low \(T\), tighter limits on the neutrino magnetic moment \(\mu_e\) can be set with a low-radioactivity detector that also has a low energy threshold. A radiopure sample of superfluid helium, scanned by infrared lasers and viewed by photodetectors, could have an energy threshold as low as 100 eV. In addition, the very good position resolution available with this technique would assist in the characterization of systematic uncertainties. Gamma ray backgrounds from Compton scattering can be partially rejected by looking for multiple scattering events in the detector volume, and x-ray backgrounds can be reduced by considering only events that occur within the central region of the detector, where x-rays are unlikely to penetrate. To avoid significant He\(_2\) diffusion, and to avoid scattering of laser light from bubbles in the liquid, this detector would be maintained at a temperature slightly below the lambda point, at roughly 2 K. The low energy threshold projected for this detector would also allow the detection of coherent neutrino-nucleus scattering \cite{27}.

Laser-induced fluorescence in superfluid helium may also be useful in the search for dark matter in the form of weakly interacting massive particles (WIMPs). The HERON group has demonstrated \cite{28} that alpha particle excitations can result in a directional roton signal in superfluid helium. This occurs because the scattering rate of rotons within the track is much higher than the scattering rate of rotons once they leave, and the rotons are more likely to be emitted perpendicular to an elongated track than parallel to it. We speculate that the trajectories of triplet molecules emerging from a spin-independent WIMP-nucleus scattering event in superfluid \(^4\)He would also show this directional effect, and thus provide information about the direction of the nuclear recoil track. Like the rotons, the diffusion of molecules within the track would be dominated by roton scattering, and the triplet molecules should be preferentially emitted perpendicular to the track. The molecule trajectories could then be determined by a laser tracking system, and the pattern of molecule trajectories could be used to determine the direction of the initial nuclear recoil. While DRIFT \cite{29} is promising for achieving directional sensitivity for WIMP-nucleus scattering events using a low-pressure time projection chamber, superfluid helium would allow a \(10^3\) times larger number density. Meyer and Sloan estimate that a nuclear recoil in \(^3\)He (from a WIMP, for example) will result in 50% of the electronic excitation that would be expected from an electron-like event for a similar energy \cite{30}. Assuming the same figure...
for $^4$He, a nuclear recoil in superfluid helium will form 6.5 $\text{He}_2(a^3\Sigma^+)$ molecules per keV, allowing a very low energy threshold. The same detector, filled with $^3$He instead of $^4$He, could also be used to place limits on spin-dependent WIMP scattering, though in this case the much lower diffusion constant would preclude determination of the nuclear recoil direction because of scattering of the molecule after it leaves the track.

Laser-induced fluorescence may also be used to detect ultracold neutrons in superfluid helium. In a currently proposed search for the neutron electric dipole moment (EDM) [4], $\text{He}_2$ singlet light emission is to be used to detect neutron absorption events in $^3$He-doped superfluid $^4$He. These events in turn monitor the precession of spin-polarized neutrons. With the addition of laser-induced fluorescence, event by event detection of both the singlet and triplet molecules and measurement of the ratio of the amplitudes of these two signals would allow the rejection of backgrounds associated with gamma ray scattering in the liquid helium and light collection optics. Due to the low temperature of the superfluid helium in the planned neutron EDM cells, the large $\text{He}_2$ diffusion constant will cause any triplet molecules to be quenched by collisions with the wall within about 3 ms. This is adequate time to cycle the triplet molecules multiple times with little effect from previous events, provided that the event rate is less than $\sim 100 \text{ s}^{-1}$. The same detection technique could also be used in experiments to measure the neutron lifetime using magnetically trapped neutrons [3].

A fascinating non-particle physics application of laser-induced fluorescence is the study of superfluid flow and turbulence. A small radioactive source, focused laser beam, or electric discharge could be used to create triplet $\text{He}_2$ molecules that would then be tracked with intersecting lasers to image their path. One approach to imaging superfluid flow is neutron absorption tomoscopy [31], which uses $^3$He as a neutral tracer and requires a finely collimated neutron beam and the ability to raster the neutron beam through the region of interest. By instead using laser induced fluorescence of $\text{He}_2(a^3\Sigma^+_u)$ molecules, many of the same superfluid helium properties can be measured, with the advantages of better position resolution and imaging in three dimensions instead of only two. Another method of imaging superfluid flow is particle image velocimetry with small (1-10 $\mu$m) neutrally buoyant glass beads [2], but this approach has so far been limited to temperatures above the lambda point.

In summary, we propose laser-induced fluorescence of metastable $\text{He}_2$ molecules as a technique for the detection of ionizing radiation with low energy threshold and good position resolution. This appears useful for applications in neutrino physics, the search for WIMPs, ultracold neutron research, and the imaging of superfluid turbulence.

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