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 He_2 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 ^{4}He.

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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 He_2 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 He_2 excimer molecules, particularly in the $He_2(A\,^1\Sigma_u^+)$ and $He_2(a\,^3\Sigma_u^+)$ 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 He_2 molecules, creating in total about 13 000 $He_2(a\,^3\Sigma_u^+)$ and 19 000 $He_2(A\,^1\Sigma_u^+)$ 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 $2\,^3$S state, which have a radiative lifetime of 8000 s in vacuum [9], only survive 15 μs in the liquid due to nonradiative bonding with ground-state atoms to form $He_2(a\,^3\Sigma_u^+)$ 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 $He_2(A\,^1\Sigma_u^+)$ has a lifetime of about 1 ns [12], while the $He_2(a\,^3\Sigma_u^+)$ 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 $He_2(a\,^3\Sigma_u^+)$ to the dissociative ground state, $He_2(X\,^1\Sigma_g^+)$, is forbidden because it requires a spin flip. A theoretical value of 18 s for the lifetime of $He_2(a\,^3\Sigma_u^+)$ 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 Z^4. 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 $He_2(a\,^3\Sigma_u^+)$ 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 ^{3}He impurities (see Fig. 1). We model the diffusion constant by the following equation:

$$\frac{1}{D} = \frac{e^{-\Delta/T}}{\Gamma_r} + \frac{T^7}{\Gamma_p} + \frac{XT^{-1/2}}{\Gamma_3} \tag{1}$$

where X is the relative concentration of ^{3}He in ^{4}He, and Δ is the roton energy gap (8.6 K). Γ_r, Γ_p, and Γ_3 represent measures of the scattering of the $He_2(a\,^3\Sigma_u^+)$ bubble with rotons, phonons, and ^{3}He. Γ_r has been determined experimentally [17] to be 2.8×10^{-6} cm^2 s^{-1} and compares well with a similar value for ^{3}He when adjusted for the effective radius [18]. A Γ_p value of 1 cm^2 s^{-1} K^7 has been determined from an experimental value for phonon-^{3}He scattering by taking into account the greater radius of the He_2 bubble [14,19], while a Γ_3 value of 3×10^{-6} cm^2 s^{-1} K$^{-1/2}$ has been similarly scaled from the experimentally determined electron mobility in dilute mixtures of ^{3}He in ^{4}He at low temperatures and correcting for the bubble size [20]. By adjusting the temperature and ^{3}He

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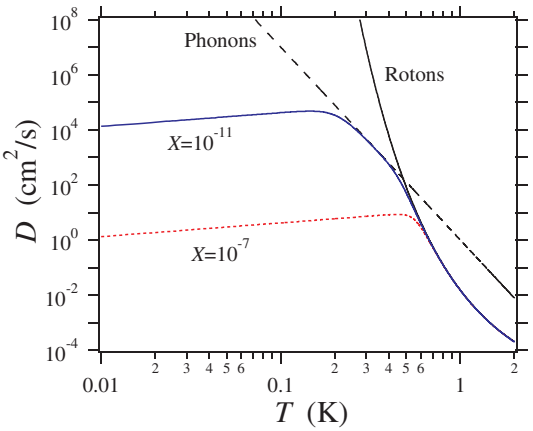

FIG. 1 (color online). Diffusion constant D versus temperature T for He_2 molecules in superfluid helium, assuming He^3 concentrations found in commercially available liquid helium ($X = 10^{-7}$) and purified liquid helium ($X = 10^{-11}$). Also shown are the individual roton and phonon contributions.

concentration, the displacement of $He_2(a\,^3\Sigma_u^+)$ over its radiative lifetime can be varied over several orders of magnitude.

Many detailed spectroscopic studies of triplet He_2 molecules in superfluid helium have been performed, and the energy levels and optical absorption frequencies are well established [7,10,21]. Figure 2 shows an energy level diagram of the lowest lying triplet states of the He_2 molecule. The arrows on the diagram show a cycling transition that can be used to detect He_2 molecules through laser-induced fluorescence. The scheme involves two pulsed infrared lasers. The first laser excites the molecule from $a\,^3\Sigma_u^+ \rightarrow c\,^3\Sigma_g^+$ with a pulse of 910 nm light. Immediately following is a second pulse to excite the molecules from $c\,^3\Sigma_g^+ \rightarrow d\,^3\Sigma_u^+$. Measured superfluid helium emission spectra indicate that this transition is at 1040 nm [10], though the absorption wavelength is expected to be blue-shifted slightly from this value [7]. We calculate that

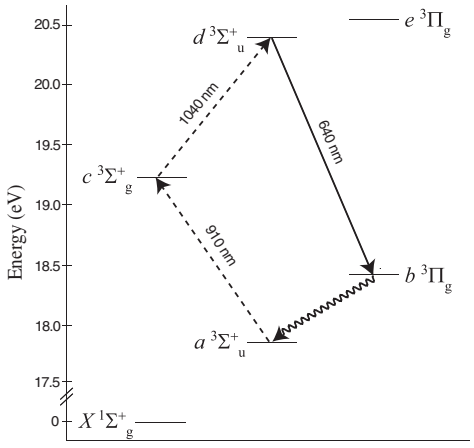

FIG. 2. Energy levels of the triplet He_2 molecule below 21 eV. Also shown is a cycling transition described in the text.

molecules in the $d\,^3\Sigma_u^+$ then decay to the $b\,^3\Pi_g$ with a 90% branching ratio, emitting an easily detected photon at 640 nm. If instead the d state decays back to the c state, it then decays back to the a state, where the molecule is again sensitive to the pump laser. The lifetime of the $d\,^3\Sigma_u^+ \rightarrow b\,^3\Pi_g$ transition has been measured to be 25 ns in both liquid [22] and gaseous [23] helium, demonstrating that there is little nonradiative quenching of the $d\,^3\Sigma_u^+$ state. Finally, the $b\,^3\Pi_g$ state is nonradiatively quenched before it can decay [7], returning to the $a\,^3\Sigma_u^+$ state where it is again sensitive to the probe laser. Thus a single triplet He_2 molecule is capable of emitting as many as 4×10^7 photons/s. Given that the cross section of the $a\,^3\Sigma_u^+ \rightarrow c\,^3\Sigma_g^+$ transition in superfluid helium is 2.4×10^{-15} cm^2 [21] and assuming a comparable cross section for the $c\,^3\Sigma_g^+ \rightarrow d\,^3\Sigma_u^+$ transition, lasers emitting 250 μJ/cm^2 pulses at each wavelength would drive the $a\,^3\Sigma_u^+ \rightarrow d\,^3\Sigma_u^+$ transition with roughly 90% efficiency per molecule. The linewidth requirement for the pump lasers is not stringent because the transitions are broadened by about 120 cm^{-1} by the liquid helium [21].

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 ± 40 ms for $a\,^3\Sigma_u^+$ molecules in superfluid helium, and they demonstrated that the rotational relaxation is much faster [21]. From the calculation of Franck-Condon factors for the cycling transition shown in Fig. 2, we have determined that only 1.2% of the molecules per cycle will fall radiatively to the first vibrational state $a(1)$ rather than the zeroth vibrational state $a(0)$. 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 at 1070 nm to 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 vibrational state where they will be blind to the pump lasers.

Alternately, intentionally driving the molecules to $a(1)$ would allow their sensitive detection using inexpensive continuous-wave (cw) diode or fiber lasers, modulated at kHz frequencies. For example, driving $a(0)$ to $c(1)$ with an 805 nm photon will cause $a(1)$ to be populated. Then, by turning off the 805 nm light and turning on a laser at 1070 nm, the molecules can be driven to $c(0)$, which will then decay to $a(0)$, emitting 910 nm photons that can be detected. Because the photon absorption cross section is

lower for transitions that change vibrational quantum number, this approach requires higher total laser power for a given cycling rate than the cycling transition described previously.

A detector using individual helium molecule fluorescence in a cycling transition would be far more sensitive to small energy depositions than a comparable scintillation detector. It should be possible to obtain a usable signal from every molecule formed in the detection region, whereas approaches using only prompt scintillation detection have to contend with incomplete detector coverage, dark counts, and the inherently low quantum efficiency of photomultipliers (typically no better than 30%). Detection of individual molecules should allow an energy threshold on the order of 100 eV for electronlike events, assuming that molecule production is linear with energy deposition. Ionization in noble gases has been shown to be roughly linear with energy deposition down to the 100 eV scale in neon and xenon gas [24], and we expect this also to be the case in helium. Use of a cycling transition allows a significant signal amplification; the total photon production is roughly 13 000 per MeV per cycle. In comparison, the radiative decay of singlet molecules in superfluid helium produces 19 000 photons per MeV.

In addition to having a very low energy threshold, a detector incorporating laser-induced fluorescence for the detection of He_2 molecules can also have very good position resolution. Because the cycling transitions described here can only produce useful fluorescence where two laser beams overlap, one can orient the beams at 90 degrees to one another and raster them throughout the volume of interest [25]. This can lead to position resolution equal to or less than the radius of the laser beam profile. Alternately, it should be possible to image tracks in the superfluid helium using an image intensifying unit or CCD.

With the properties mentioned above, a detector using superfluid helium could be used in several different configurations. One mode of operation is a simple continuous scan of the detector volume to detect energy deposition events. A second is to use a layer of wavelength shifting fluor [26] to convert the singlet He_2 scintillation light to the visible. The visible light would be detected by photomultipliers, triggering the laser scanning system. The lasers need only be activated following event detection, and some particle identification should be possible by measuring the ratio of prompt scintillation light to laser-induced fluorescence. In other liquified noble gases, this ratio has been found to vary with excitation type [27], which is likely the case in liquid helium as well [8]. In addition, requiring the detection of both prompt scintillation light and laser-induced fluorescence would allow the rejection of backgrounds due to any interactions that do not take place within the liquid helium. The disadvantage of this second mode is that the energy threshold will be set by the efficiency of detecting the prompt scintillation light, while in the first mode the energy threshold is set by the amplified triplet molecule signal. Depending on the event rate of the ionizing particles, the triplet molecules can represent a considerable background due to their long lifetime. For experiments with high event rates, this background can be reduced by lowering the helium temperature, thereby increasing the mobility of the triplet molecules and lowering their lifetime. This has the disadvantage of decreasing the number of times each molecule can be cycled.

The projected low energy threshold provides a unique opportunity to search for the neutrino magnetic moment. Because neutrino-electron scattering is enhanced for low electron recoil energies, tighter limits on the neutrino magnetic moment can be set with a low-radioactivity detector that also has a low energy threshold [28]. A radiopure sample of superfluid helium, scanned by infrared lasers and viewed by photodetectors, could have an energy threshold as low as 100 eV. γ-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 [29].

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 [30] 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. 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 electronlike event for a similar energy [31]. Assuming the same figure for ^{4}He, a nuclear recoil in superfluid helium will form 6.5 $He_2(a\,^3\Sigma_u^+)$ 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 [4], 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 γ-ray scattering in the liquid helium and light collection optics. Because of the low temperature of the superfluid helium in the planned neutron electric dipole moment cells, the large 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 $\sim 100\ s^{-1}$. The same detection technique could also be used in experiments to measure the neutron lifetime using magnetically trapped neutrons [3].

A fascinating nonparticle 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 He_2 molecules that would then be tracked with intersecting lasers to image their path. One approach to imaging superfluid flow is neutron absorption tomography [32], 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 $He_2(a\,^3\Sigma_u^+)$ 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 μm) neutrally buoyant glass beads [33], but this approach has so far been limited to temperatures above the lambda point.

In summary, we propose laser-induced fluorescence of metastable 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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