The Importance of Nonlinear H$_2$ Photoexcitation in Strongly
Irradiated PDRs

P.P Sorokin and J.H. Glownia
IBM Research Division
P.O. Box 218, Yorktown Heights, NY 10598-0218
sorokin@us.ibm.com; glownia@us.ibm.com

and

R.T. Hodgson
822 Pinesbridge Rd., Ossining, NY 10562
patents@aip.org

Received ___________; accepted ___________
ABSTRACT

It is shown that, under sufficiently intense OB-star illumination of a stationary photoexcitation front (PDR), nonlinear H$_2$ photoexcitation processes comprising driven resonant two-photon transitions between X-state quantum levels, with VUV continuum light from the star supplying both driving fields, largely determine the photonic pathways of H$_2$ molecules in the PDR close to the ionization front. Specifically, for a flux of $\sim 4 \times 10^5$ Habing fields incident upon a PDR, the total rate at which an H$_2$ molecule is nonlinearly photoexcited out of any X-state quantum level is calculated to be roughly 100 times greater than the total rate at which it is linearly photoexcited out of the same level. In strongly excited PDRs, the populations in almost all of the $\sim 300$ bound quantum levels of the X state will be maintained approximately equal via a few myriads of interconnecting two-photon steps. The remarkable importance of two-photon transitions in H$_2$ photoexcitation in strongly irradiated PDRs derives from the exceptionally narrow Raman linewidth ($\Gamma \sim 10^{-6}$ sec$^{-1}$) that characterizes all two-photon transitions between bound H$_2$ X-state quantum levels.

Subject headings: infrared: ISM - - - ISM: clouds - - - ISM: HII regions - - - ISM: lines - - - ISM: lines and bands - - - ultraviolet: ISM
1. Introduction

In existing theoretical models of stationary photoexcitation fronts (PDRs) [see Draine & Bertoldi (1996) for a comprehensive list of references], a one-dimensional geometry is usually considered, with light from an OB-type illuminating star assumed incident upon the PDR from one direction. In all published PDR models, only linear H$_2$ photoexcitation processes have been considered. The consensus among astrophysicists is that these models adequately describe the main photonic events that occur in H$_2$-containing clouds separated by at least 10 pc from OB-type illuminating stars. In the present Letter, we draw attention to the fact that, in very strongly irradiated PDRs (for example, those with cloud-to-star distances $\leq 0.1$ pc), nonlinear photoexcitation of H$_2$ can evidently become the dominant photonic process determining the structure of the PDR.

2. Resonantly Enhanced, VUV Starlight-Driven, Two-Photon Transitions between H$_2$ X-State Quantum Levels

An estimate of the resonantly-enhanced, VUV starlight-driven, two-photon transition rate between two H$_2$ X-state quantum levels can be obtained with use of the formula shown in Fig. 1. In SI units, this formula [c.f. eq. (5.6b) of Hanna et al. (1979)] gives the cross-section for Stokes-wave gain in resonant stimulated Raman scattering (SRS) in a three-level system. However, it equally well gives the cross-section for induced nonlinear absorption at $\omega_P$ when an intense radiation field is present at $\omega_S$ - a process often referred to as inverse Raman scattering (IRS). It has long been recognized (Jone & Stoicheff 1964; MacQuillan & Stoicheff 1966) that both processes are fundamentally the same. In this equation, the classical electron radius $r_e \approx 2.82 \times 10^{-15}$ m. The quantity $I_P$ is the pump intensity in W/m$^2$. The subscripts $g$, $i$, and $f$ designate the ground level, intermediate level, and final level, respectively, of the three-level system. The quantity $\Gamma$ is the Raman linewidth (in units of angular frequency), which, for the astrophysical environment assumed in our PDR model, would be entirely determined by a convolution of the vibrational decay rates of the two X$n''$ quantum levels involved. In light of recent calculations of quadrupole transition probabilities for all X-state radiative decays (Wolniewicz et al. 1998), one can realistically assume a value $\Gamma = 10^{-6}$ sec$^{-1}$ for all two-photon transitions between X-state quantum levels. The quantities $f_{fi}$ and $f_{gi}$ are the oscillator strengths of the Stokes-wave and primary frequency transitions. For all H$_2$ Lyman (B$\leftarrow X$) and Werner (C$\leftarrow X$) transitions, these $f$-values can be simply determined from the tables in Abgrall et al. (1993 a,b). The quantities such as $\Omega_{if}$ are the energy separations of the levels involved, expressed as angular frequencies, e.g. $(\hbar/2\pi)\Omega_{if} = E_i - E_f$. Note that the Stokes-wave frequency $\omega_S$ essentially cancels $\Omega_{if}$ in the present case. The equation in Fig. 1 gives the cross-section in units of m$^2$.

To estimate the rates of both nonlinear and linear H$_2$ photoexcitation in the PDR, one needs to know the VUV flux from the star incident upon the cloud surface. Here, for definiteness, we assume the illuminating star to be a B0 III star, with temperature $T=31,500$K and radius $R = 16\,\odot$. We take the cloud-to-star distance to be 0.1 pc, and assume that there is no intervening dust cloud between the star and H$_2$-containing cloud. From the Planck formula for emittance of photons from a blackbody per cm$^2$ per sec per unit frequency range (Allen 1976), one finds that at 1,000 Å the incident flux on the PDR is $\phi_{1000} = 2.55 \times 10^8$ photons per cm$^2$ per sec per cm$^{-1}$. Habing (1968) estimated the intensity
of interstellar starlight at $\lambda = 1,000 \, \text{Å}$ to be $\lambda u_{\lambda} = 4 \times 10^{-14} \, \text{erg cm}^{-3}$. At the surface of the PDR we are here considering, therefore, $\phi_{1000}$ is about $4.25 \times 10^{5}$ Habing fields.

As is the case in almost all published PDR models, it is here assumed that ionizing radiation (i.e. $\lambda \leq 912 \, \text{Å}$) from the star creates a thin H II region on the surface of the PDR cloud nearest the star. Within this H II region, a large fraction ($\sim 2/3$) of the incident ionizing photons are converted to Ly-$\alpha$ photons via $H^+ + e^- \rightarrow$ recombinations followed by cascading transitions through H-atom excited states. Half of the Ly-$\alpha$ photons enter the neutral region, with their frequencies having become distributed in a $\sim 20$-cm$^{-1}$-wide spectral band via the frequency redistribution that results from elastic scattering by hot H atoms in the H II region. One can easily estimate the total flux of ionizing photons incident upon the PDR from tables of the Planck function (Allen 1976). One finds this to be $\phi_{\text{ion}} \approx 6.2 \times 10^{12}$ photons cm$^{-2}$ sec$^{-1}$. There are thus about $\phi_{\text{Ly}\alpha} \approx 2.1 \times 10^{12}$ Ly-$\alpha$ photons entering the neutral PDR region per cm$^2$ per second. Thus, the Ly-$\alpha$ flux (per unit wavenumber) entering the neutral PDR region is seen to exceed that at 1,000 Å by roughly a factor 400. In §3, we will briefly hint at an important effect this Ly-$\alpha$ radiation should have on the nonlinear excitation of $H_2$ in strongly irradiated PDRs.

To estimate the total rate at which an $H_2$ molecule in the PDR would undergo linear photoexcitation, one can use Table 2 of Draine & Bertoldi (1996). These authors have calculated and summed individual contributions to the total unshielded $H_2$ linear photoexcitation rate from all $X \rightarrow B$ and $X \rightarrow C$ transitions. For a flux $\phi_{1000} = 4.25 \times 10^{5}$ Habing fields, the total linear photoexcitation rate for a molecule in $(X_0, J'' = 1)$ would be about $1.2 \times 10^{-4}$ sec$^{-1}$. In the PDR model of Draine & Bertoldi (1996), a Doppler width $\approx 1.7$ cm$^{-1}$ is assumed.

With use of the equation shown in Fig. 1, we now evaluate the driven (simultaneous, not sequential) two-photon transition rate from $X_0, J'' = 1$ to $X_1, J'' = 1$ involving $C0(\Pi_u^-$), $J' = 1$ as resonant intermediate state. A rough - but conservative - approximation one can make in integrating the expression for $\sigma_{\text{IRS}}$ over the applied VUV pumping continuum is to assign to the quantity $(\Omega_{ig} - \omega_P)$ appearing in the denominator of the equation in Fig. 1 a constant value corresponding to an offset of one wavenumber, and then integrate over a spectral interval two wavenumbers wide on either side of the resonance. From the value of $\phi_{1000}$ earlier estimated, the value of $I_P$ for a four-wavenumbers-wide spectral interval is found to be $0.00002$ W/m$^2$. With substitution of appropriate values for the other quantities appearing in the equation of Fig. 1 (e.g. $f_{gi} = 0.03$, $f_{ji} = 0.07$), one finds the cross-section for induced absorption to be $\sigma_{\text{IRS}} \approx 8.4 \times 10^{-14}$ cm$^2$. Multiplying this value by $\phi_{1000} \times 4$ shows the transition rate of the IRS process to be $\sim 8.6 \times 10^{-5}$ sec$^{-1}$. The rate of nonlinear photoexcitation of an $H_2$ molecule in $(X_0, J'' = 1)$ via this particular two-photon transition is thus about 0.7 times the earlier estimated total rate of linear photoexcitation of an $H_2$ molecule in the same $(X_0, J'' = 1)$ quantum level.

However, driven two-photon transitions from $(X_0, J'' = 1)$ to $(X_1, J'' = 1)$ can also occur via the paired transitions $Cn-0Q1$, $Cn-1Q1$ ($n = 1-5$) and also $Cn-0R1$, $Cn-1R1$ ($n = 0-5$). This makes the total two-photon transition rate out of $(X_0, J'' = 1)$ exceed the total linear rate by $\sim 7$ times. (The wavelengths of both $C6-0Q1$ and $C6-0R1$ are shorter than 912 Å, the Lyman limit. No $\lambda \leq 912$ Å starlight can penetrate beyond the HII region of the PDR.) One can also go from $(X_0, J'' = 1)$ to $(X_1, J'' = 3)$ via transitions $Cn-0R1$, $Cn-1P3$. With the inclusion of these paths, the two-photon transition rate out of $(X_0, J'' = 1)$ would be roughly 10 times the total linear rate of photoexcitation. One next must consider
analagous two-photon transitions from \((X_0, J''=1)\) that terminate on \((X_2, J''=1), (X_2, J''=3), \ldots (X_8, J''=1), (X_8, J''=3), \ldots\). With inclusion of these, the two-photon excitation rate from \((X_0, J''=1)\) would exceed the total linear excitation rate by \(\sim 70\) times. Finally, two-photon excitation from \((X_0, J''=1)\) can also occur via B-state resonant intermediate levels. Additional inclusion of such pathways, makes the total rate of two-photon excitation out of \((X_0, J''=1)\) exceed the total linear photoexcitation rate out of the same quantum level by at least a factor 100. Roughly the same nonlinear-to-linear photoexcitation rate ratio would obtain for \(\text{H}_2\) molecules in any of the \(\approx 300\) X-state quantum levels. The estimated total nonlinear photoexcitation rate out of any such level is thus \(\sim 1.2 \times 10^{-2}\) sec\(^{-1}\).

3. Consequences of VUV Starlight-Driven Two-Photon Transitions Occurring in Heavily Irradiated PDRs

From the transition rates calculated above, it is apparent that, through a few myriads of interconnecting two-photon transitions, the \(\text{H}_2\) populations in each of the \(\approx 300\) X-state quantum levels would be maintained approximately equal. Note that no \(\text{H}_2\) photodissociation can result from two-photon transitions occurring from the highest bound X-state levels to unbound X-state continuum levels. The two-photon transition rate depends inversely upon the Raman linewidth, and the latter represents a convolution of the lifetimes of the quantum levels connected by the two-photon process. When one steps from the highest bound X-state levels to unbound levels of the X-state continuum, the lifetime drops by roughly eighteen orders of magnitude! On the basis of this same disparity in lifetimes, one perhaps should question the validity of the assumption usually made in linear PDR models that roughly one out of ten \(\text{H}_2\) photoexcitations from X-state quantum levels result in photodissociation via fluorescent VUV transitions terminating on unbound levels of the X-state. This would be true if linear photoexcitation of \(\text{H}_2\) in PDRs did involve real excitation of B- and C-state quantum levels. However, in collisionless media, it is known (Loudon 1983) that linear photoexcitation occurs entirely via spontaneous resonant Raman scattering, and, at exact resonance, the cross-section for the latter varies again as \(\Gamma^{-1}\) (Shen 1974).

If one knows the total \(\text{H}_2\) density in the neutral region of the PDR, one can approximately estimate the maximum thickness \(d\) of the nonlinearly excited region as follows. Assume an \(\text{H}_2\) density of \(10^4\) cm\(^{-3}\). In the nonlinearly excited region, these molecules will be evenly distributed amongst all the \(\approx 300\) X-state quantum levels, with the spontaneous infrared radiation emission rate from each level being \(\sim 10^{-6}\) sec\(^{-1}\). Assume the average IR emitted photon to have an energy of \(5,000\) cm\(^{-1}\). Equating the VUV power from the star incident upon the PDR in the entire spectral range of the Lyman and Werner bands that lies longward of 912 \(\AA\) to the IR power radiated in a column of thickness \(d\):

\[
(20,000)(2.55 \times 10^8)(100,000) \approx (10^4)(10^{-6})(5,000)d,
\]

one finds \(d\approx 0.003\) pc. Figure 1 of [Lemaire et al. (1996)] is a striking colored image (colors corresponding to emission intensity) of \(\text{H}_2\) 1-0 S(1) vibrational emission at 2.121 \(\mu\)m in NGC 7023, taken at very high spatial resolution. It shows \(\text{H}_2\) vibrational emission originating from a \(\sim 0.02\)-pc-thick, roughly planar, cloud located \(\sim 0.07\) pc to the NW of the illuminating star HD 200775. (From Earth, one views this cloud largely through one of its 'edges'.) There is very much higher \(\text{H}_2\) emission coming from three \(\sim 0.004\)-pc-thick embedded filaments located near the surface of the cloud facing the star. In view of the
calculations that have been presented above, we suggest that these bright filaments in NGC 7023 might represent PDR regions that are nonlinearly excited.

It was noted above that Ly-α radiation generated in the H II region enters the PDR neutral region at an intensity level roughly 400 times greater than the continuum level at 1,000 Å, but only over an estimated ~20-cm⁻¹-wide bandwidth. There are a dozen or so allowed transitions originating from X-state quantum levels whose frequencies fall within such a Ly-α spectral distribution. A slight contribution towards the equalization of H₂ populations in the various X-state quantum levels arises from these resonances. However, there is a different - rather profound - effect that this Ly-α radiation can induce, when it is applied to an equally populated manifold of X-state quantum levels. It can induce broadband stimulated Raman scattering (SRS) to occur on the three or four strongest of the above mentioned resonant transitions, generating broadband IR Stokes-wave light on strong transitions to EF-states. An SRS process would occur as part of a 2n-wave parametric oscillation (SRS-PO) process, with additional IR and VUV light being generated on strong transitions ultimately returning molecules to the X-state levels from which the SRS-PO processes originated. As shown elsewhere (Sorokin & Glownia 2001), the so-called ‘unidentified infrared (emission) bands (UIBs)’ can be successfully assigned to IR emissions coherently generated in such SRS-PO processes.

One of the authors (PPS) acknowledges receiving strong encouragement from Dr. Anita J. Schell-Sorokin to publish the central idea upon which this article is based.
REFERENCES

Abgrall, H., Roueff, E., Launay, F., Roncin, J.-Y., & Subtil, J.-L. 1993a, A&AS, 101, 273.
———. 1993b, A&AS, 101, 323.

Allen, C.W. 1976, Astrophysical Quantities (London and Dover, New Hampshire: The Athlone Press).

Draine, B.T. & Bertoldi, F. 1996, ApJ, 468, 269.

Habing, H. J. 1968, Bull. Astron. Inst. Netherlands, 19, 421.

Hanna, D.C., Yuratich, M.A., & Cotter, D. 1979, Nonlinear Optics of Free Atoms and Molecules (Berlin, Heidelberg, & New York: Springer-Verlag).

Jones, W.J. & Stoicheff, B.P. 1964, Phys. Rev. Lett., 13, 657.

Lemaire, J.L., Field, D., Gerin, M., Leach, S., Pineau des Forêts, G., Rostas, F., & Rouan, D. 1996, A&A, 308, 895.

Loudon, R. 1983, The Quantum Theory of Light (2d ed., Oxford: Clarendon Press).

MacQuillan, A.K. & Stoicheff, B.P. 1966, in Physics of Quantum Electronics, ed. P.L. Kelley, B. Lax, & P.E. Tannenwald (New York: McGraw-Hill), 192.

Shen, Y.R. 1974, Phys. Rev. B, 9, 622.

Sorokin, P.P. & Glownia, J.H. 2001, to be published.

Wolniewicz, L., Simbotin, I., & Dalgarno, A. 1998, ApJS, 115, 293.

This manuscript was prepared with the AAS LATEX macros v4.0.
\[ \sigma_{IRS} = \frac{4\pi^3 r_e^2 c^2}{h} \frac{\omega_s}{\Gamma} \left( \frac{f_{fi}}{\Omega_{if}} \right) \left( \frac{f_{gi}}{\Omega_{ig}} \right) \left( \frac{I_p}{(\Omega_{ig} - \omega_P)^2} \right) \]

Fig. 1.— Equation for the cross-section of either SRS Stokes-wave gain or IRS-induced absorption in a resonant, three-level system. The light waves in the diagram are depicted for the SRS case.