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TITLE **PRESSURE BROADENING OF THE $[(dt\mu)dee]^*$ FORMATION RESONANCES**

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ABSTRACT

The treatment of $[(dt\mu)dee]^*$ formation at high densities as a pressure broadening process is discussed. The quasistatic approximation is shown to satisfy the usual conditions of muon-catalyzed fusion better than does the impact approximation. Complete accurate results are shown for the impact approximation, and a preliminary rough treatment is presented to illustrate the quasistatic approximation.

I. INTRODUCTION

The diagram in Fig. 1 shows the currently predicted positions[1] of the resonances that may contribute to $dt\mu$ molecular formation (mf) in a collision of $t\mu$ with D_2 at low temperature,

$$t\mu + D_2\,[\nu_i=0,\ J_i] \longrightarrow (dt\mu)dee\,[\nu_f=2,\ J_f]\ . \tag{1}$$

In all cases considered here the target D_2 is in its ground vibrational state and the complex $(dt\mu)dee$ is formed with electronic molecular vibrational quantum number of 2, so the various possible transitions will be designated by the initial and final rotational quantum numbers, $J_i \rightarrow J_f$. The amplitude shown for each resonance in Fig. 1 is *roughly* proportional to its mf matrix element *and* to the abundance of the initial state in a low-temperature target. Until Petrov[2] published his germinative paper in 1985, each of these resonances was viewed as a δ function.[3] The δ functions above threshold can be reached by energetic $t\mu$ atoms in the Maxwellian distribution, but those below threshold are completely inaccessible in this picture. Petrov[2] pointed out that each resonance actually has a finite width due both to intramolecular (electronic Auger) contributions as well as intermolecular (collisional) effects. Menshikov and Ponomarev[4] called attention to the possibility that three-body effects,

$$t\mu + D_2 + X \longrightarrow (dt\mu)dee + X \tag{2}$$

where X is D_2, DT, or T_2, could be responsible for the observation of Jones *et al.*[5] of a nonlinear dependence of the mf rate on density. We now believe that these three (*or more*) body effects are very usefully interpreted as pressure broadening of the resonances.

II. PRESSURE BROADENING APPROXIMATIONS

In the diagram of Fig. 2, we have picked out the two most promising below-threshold resonances and attached a *hypothetical* line-shape function to them. The contribution of a negative-energy resonance to the mf rate is given by the overlap of the line profile with the thermal (e.g., Maxwellian) distribution — this occurs, of course, only at positive energies. Now we have drawn

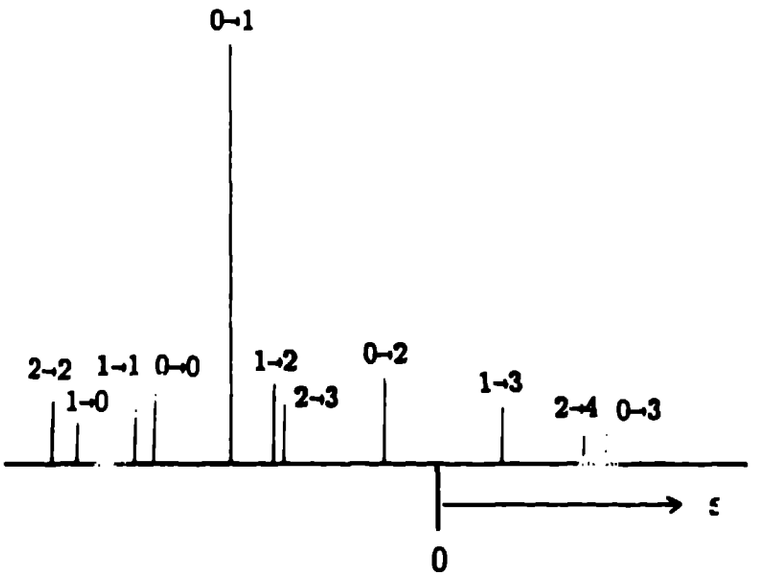

Fig. 1. Schematic of $[(dt\mu)dee]^*$ formation resonances, $J_i \rightarrow J_f$, with heights roughly indicative of the size of the matrix element and abundance of the initial state in a low-temperature target.

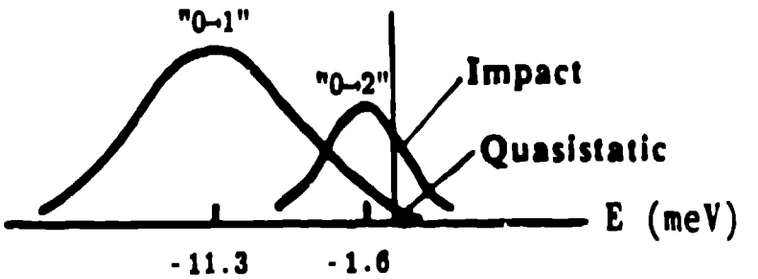

Fig. 2. The two most important below-threshold resonances. The line shapes here are drawn to exhibit the impact and quasistatic approximations and have no quantitative significance.

these two profiles as if one, the 0→2 resonance, lies very close to threshold, whereas the other, the 0→1 resonance, reaches threshold only with its far wing. This situation illustrates, though not completely as we shall see, the two main line-broadening approximations: the impact approximation valid for small $|\Delta E|$ and the quasistatic approximation valid for large $|\Delta E|$.[6] We will say more in Sec. III about the validity of these two mutually exclusive approximations, but for now we just want to show what is needed to carry them out. The most used approximation in muon-catalyzed fusion (μCF) in the past has been the impact approximation, sometimes called the Lorentzian approximation because it always yields a line shape with Lorentzian functional form,

$$I_{\rm imp}(\omega) = \frac{\Gamma/\pi}{(\omega-\omega_0)^2 + (\Gamma/2)^2} \ . \tag{3}$$

There are two formulas for the width Γ of the Lorentzian, the usual one from optics[6-7] and one recently proposed by Menshikov.[8] In both, the width is simply proportional to the density. In optics where the photon carries negligible momentum, Γ depends on the incoherent sum of the inelastic cross sections for the initial *and* final states plus the coherent difference of the elastic scattering amplitudes,[7]

$$\Gamma_{\rm op} = \hbar \langle\, nv[\, \sigma_{\rm i}^{\rm inel} + \sigma_{\rm f}^{\rm inel} + \iint \sin\theta \ d\theta\, d\phi\, |f_{\rm i}^{\rm l}(\theta,\phi) - f_{\rm f}^{\rm l}(\theta,\phi)|^2] \,\rangle_{\rm T} \ , \tag{4}$$

where the indicated average is over the velocity distribution and n is the density.

However, when the photon is replaced by the massive $t\mu$ atom, Menshikov[8] has asserted that the impact of $t\mu$ completely disrupts the effect of the initial state and the result is the same as Eq. (4) *except* with the initial state deleted; i.e., it is given by the total cross section for the final state only,

$$\Gamma_{\rm sr} = \hbar \langle\, nv[\, \sigma_{\rm f}^{\rm inel} + \sigma_{\rm f}^{\rm el}] \,\rangle_{\rm T} \tag{5}$$

where "sr" stands for strong recoil. We have accurately calculated all cross sections needed in either case, and the results will be given in Sec. IV.

The *opposite* approximation is known as the quasistatic approximation.[6] It depends on the wave function *during* the collisions, not just on asymptotic properties like cross sections. In the usual quasistatic formula,

$$I_{\rm qs}(\omega) = \int \rho(\vec{R})\, \delta[\, V_{\rm f}(\vec{R}) - V_{\rm i}(\vec{R}) \quad \hbar\omega \,]\, d^3R \ . \tag{6}$$

Here $\rho(\vec{r})$ is the spatial distribution of all molecules, which depends on the system potential energy and the temperature, and serves in Eq. (6) as the weighting function of the energy shift of the target molecule due to the different potential it sees before and after the transition. Equation (6) does not yield a universal line-shape function analogous to Eq. (3), however, the shape is generally exponential in the line wings. Also, the quasistatic width is not simply proportional to the density like the impact width

The pressure-broadened mf rate is usually written

$$\lambda_{mf}^{i}(T) = \int_0^\infty d\epsilon \;\; \lambda_{mf}^{(2)i}(\epsilon, T)\; I(\epsilon - \epsilon_i, T) \tag{7}$$

where ϵ_i is the unperturbed energy of the i^{th} resonance, $\lambda_{mf}^{(2)i}(\epsilon, T)$ is the two-body rate at the perturbed energy ϵ, and $I(\epsilon - \epsilon_i, T)$ is the line-broadening function. It is usually assumed that the broadening function does not depend in any essential way on the particular resonance being considered. Now through this assumption is intuitively appealing and operationally convenient, its validity is not really obvious. First of all, the cross sections for collisions with the bath molecules depend on the quantum numbers of the molecule, but this dependence has been shown to be relatively weak.[9-10] Possibly more important is the dependence that comes about because the center of mass of $(dt\mu)dee$ does not coincide with that of the D_2 molecule from which it is formed.[11] This shift causes the broadening to depend on the angular momentum of the transition; i.e., the broadening is essentially different for the 0→1 and 0→2 transitions. Numerically the difference could be as much as a factor of 2 for a given $\Delta E\,(=\epsilon - \epsilon_i)$. Of course, as a center-of-mass effect it can only change things to the extent that the bath molecules alter the recoil energy. The actual importance of this observation is not yet known.

III. CRITERIA FOR LINE-BROADENING APPROXIMATIONS

The fundamental conditions for the impact approximation come from the requirement that the Fourier integral (note $\Delta E = \hbar\omega$)

$$I(\omega) = \frac{1}{\pi}\mathcal{R}e\left[\int_0^\infty \Phi(t)\; e^{-i\omega t}\; dt\right] \tag{8}$$

of the correlation function

$$\Phi(t) = \langle\, f^*(0)\, f(t)\,\rangle_T \tag{9}$$

yield a Lorentzian.[6] $\Phi(t)$ is the ensemble average of the overlap of the functional f describing the oscillation of the system at different times, subject to the interactions with the neighboring bath molecules. For Eq. (8) to yield a Lorentzian, $\Phi(t)$ must be an exponential at times that contribute most to the integral; i.e., the correlation function must be exponentially decaying by times $t \sim |\omega|^{-1}$. This condition requires, first of all, binary collisions since otherwise the interaction is maintained by additional collisions and can never exponentially decay. Secondly, the detuning $|\omega|$ must not be too large since otherwise the interaction will still be dynamically developing during the transition. The first condition is given by the inequality

$$\Delta t_c >> \tau_c \tag{10}$$

where

$$\Delta t_c = \frac{1}{n v \sigma} \tag{11}$$

is the time *between* collisions and

$$\tau_c = \frac{\rho_c}{v} \simeq \frac{1}{v}\left[\frac{\sigma}{\pi}\right]^{\frac{1}{2}} \tag{12}$$

is the *duration* of a collision. The second condition is given by the inequality

$$|\omega| << \tau_c^{-1} \ . \tag{13}$$

Note that a different criterion has often been stated to justify the application of the impact approximation to $dt\mu$ formation, namely,

$$|\Delta E| \lesssim \Gamma \ . \tag{14}$$

To see how this inequality is related to the *fundamental* conditions above, Eqs. (10) and (13), we can use the relation

$$\Gamma \simeq \frac{\hbar}{\Delta t_c} \ . \tag{15}$$

Hence we can rewrite Eq. (10) as

$$\tau_c << \Gamma/\hbar \tag{16}$$

and Eq. (13) as

$$\tau_c << |\Delta E|/\hbar \ ; \tag{17}$$

however, these two inequalities imply nothing about the relation of Γ to $|\Delta E|$ and, in fact, Eq. (14) is neither a necessary nor a sufficient condition for the impact approximation.

Now we will evaluate Eqs. (10) and (13) for d–t μCF conditions. For this purpose it is convenient to rewrite the conditions as

$$\phi << \frac{\sqrt{\pi}}{n_0 \sigma^{3/2}} = \frac{8.3 \times 10^{-23}}{\sigma^{3/2}} \ , \tag{10'}$$

where n_0 is the density of liquid hydrogen (LHD) and ϕ is the target density in LHD units, and

$$|\Delta E| << \hbar v \sqrt{\pi}/\bar{v} = 1.17 \times 10^{-12}\, v/\sqrt{\sigma} \ \text{meV} \ . \tag{13'}$$

At 300° K, $v \simeq 1.6 \times 10^5$ cm/s and $\sigma \simeq 7 \times 10^{-15}$ cm^2, yielding conditions

$$\phi << 0.14$$

and

$$|\Delta E| << 2.2 \text{ meV}$$

for validity of the impact approximation. At 30°K, the velocity, $v \simeq 5.0 \times 10^4$ cm/s, is lower and the cross section, $\sigma \simeq 2.5 \times 10^{-14}$ cm^2, is larger, so the situation is even worse for the impact approximation,

$$\phi << 0.02$$

and

$$|\Delta E| << 0.4 \text{ meV}$$

being required. Clearly, the impact approximation is not valid for the usual experimental target condition of near-liquid density. It will also *never* be valid for the 0→1 resonance if that resonance lies ~11 meV below threshold as predicted.

The quasistatic approximation is valid if the inequality of *either* Eq. (10) or (13) is reversed. Generally a many-body, rather than a binary quasistatic calculation, will be required at high densities; however, a binary approximation may still be valid in the far wing of a line. To the extent that the bath molecules alter the target recoil from the $t\mu$ impact there may be an additional kinetic-energy effect. Hence the criteria for validity of Eq. (6) may be somewhat more stringent than in optics.

IV. RESULTS IN THE IMPACT APPROXIMATION

We have calculated elastic[10] and rotationally inelastic[9] cross sections using the quantum mechanical close-coupling method for collisions of $(dt\mu)dee$ and $(dd\mu)dee$ with normal molecules in the μCF targets. Some of the inelastic cross sections contribute to stabilization of the resonant complex initially formed; they are substantially smaller than the cross sections obtained by Ostrovsky and Ustimov[12] using a scaling law derived in the Born approximation. For example, the rate for the $J=0 \rightarrow J=1$ transition in $(dt\mu)dee + D_2$ collisions at 100°K is about 6 times smaller than calculated by Ostrovsky and Ustimov. However, the present interest is in the total broadening of the resonance due to these collisions. Both the elastic and the inelastic cross sections are required in the impact-approximation formulas (4) and (5). The calculations of these quantities have been described in detail previously,[9-10] so the important results will just be summarized here.

The cross sections contributing to the impact broadening of reaction (1) are shown in Fig. 3. The broadening is clearly dominated by elastic scattering whether the coherence difference, required for Eq. (4), or just the final-state $[(dt\mu)dee]$ cross section, required for Eq. (5), is used. Next in importance are the rotationally inelastic cross sections of $(dt\mu)dee$. These cross sections are dominated by $\Delta J=1$ transitions that are made possible by the unequal masses of $dt\mu$ and d. In D_2 only transitions with ΔJ *even* are allowed so the corresponding inelastic cross section is much smaller. Although the inelastic cross sections are increasing while the elastic cross sections are decreasing as the collision energy increases, elastic scattering is still the dominant contributor to the line broadening at 100 meV. The reorientation (m changing) cross sections are unimportant at all energies.

The above cross sections were actually calculated for $J_i=0$ and $J_f=1$, but

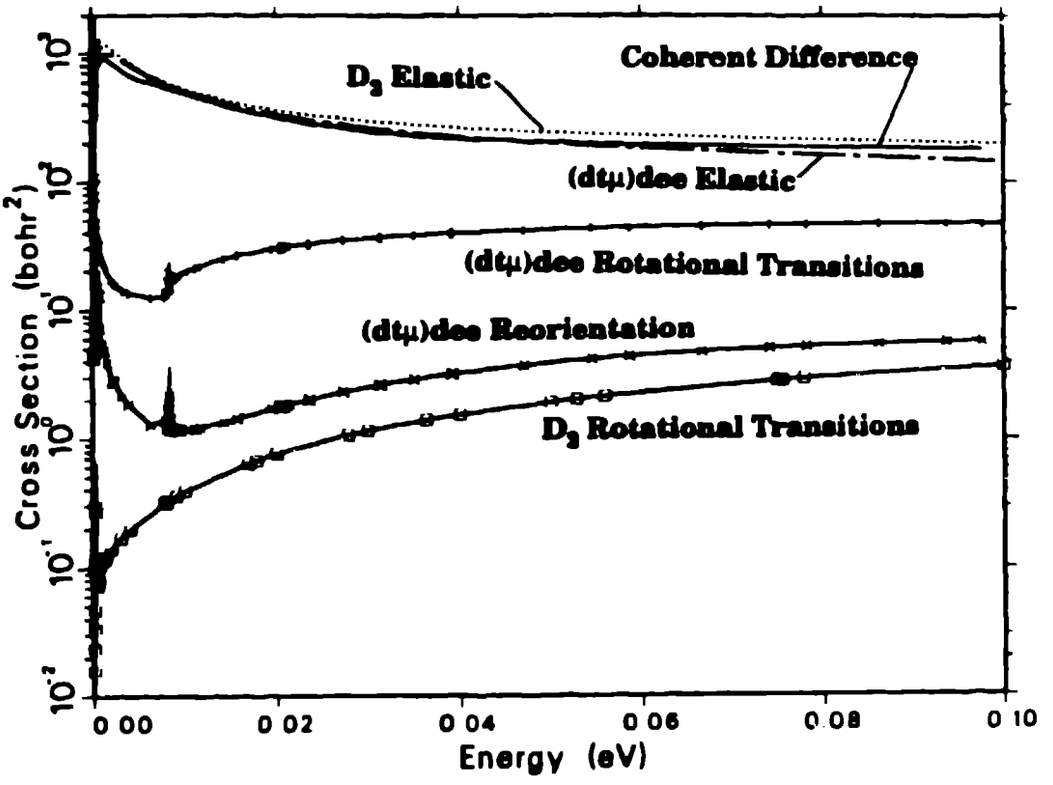

Fig. 3. Cross sections for calculation of the resonance width in the impact approximation for $t\mu + D_2 + D_2 \longrightarrow (dt\mu)dee + D_2$.

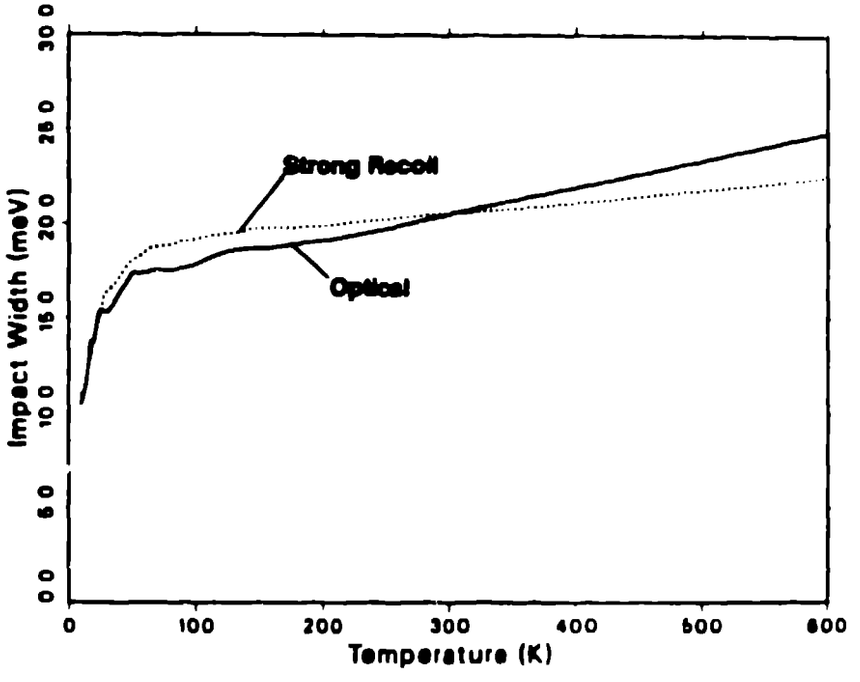

Fig. 4. Impact-approximation widths at LHD. The solid curve is calculated with Eq. (4), the dashed curve with Eq. (5).

they are rather insensitive to these choices. Hence it is reasonable to use the same cross sections for other J_i and J_f. The results of using these cross sections in Eqs. (4) and (5) are shown in Fig. 4. Remarkably, the calculated widths are about the same in the optical and strong-recoil formulations, and hence the impact analysis of line broadening will be insensitive to this choice. The width is more than three times that previously calculated[8] for a temperature of 23°K. However, the very restrictive conditions for applicability of the impact approximation to pressure broadening of the $(dt\mu)dee$ formation resonances should be kept in mind.

Disregarding this caution for the moment, we show in Fig. 5 the mf rates as a function of density for the 0→1 and 0→2 resonances using the impact-approximation width at 20°K. At this temperature the impact width is about 14ϕ meV. In this approximation the mf matrix element is clearly more important than the detuning; i.e., the 0→1 resonance at 11 meV below threshold is still more important than the 0→2 resonance at only 1.6 meV below threshold. In Fig. 6 the effect of raising or lowering the 0→2 resonance energy by 2 meV is shown. The former moves the resonance above threshold. The actual resonance energies are uncertain by at least this amount. As can also be seen, an increase in the width (e.g., with ϕ) when the resonance is very close to threshold can actually decrease the mf rate; i.e., there exists an optimum width that maximizes the overlap with the thermal distribution.

V. QUASISTATIC APPROXIMATION

An accurate quasistatic calculation of the pressure broadening of the $(dt\mu)dee$ formation resonance would require a complete potential surface including the dependence on the intramolecular vibrational coordinate. Such a potential surface is not yet available, but it still seems desirable to exhibit the qualitative behavior expected. For this purpose we have carried out a *rough* quasistatic calculation assuming:

1. Binary interactions
2. Exponential intermolecular potential
3. Interaction primarily due to nearest atoms (treating $dt\mu$ as a nucleus)
4. Rapid vibration and slow rotation compared with thermal motions.

The distribution of distances between nearest particles is given by

$$\rho(R)\, dR \propto 4\pi R^2 n \exp\left[-\frac{4\pi}{3} nR^3 - \frac{V(R)}{kT}\right] dR \tag{18}$$

using the potential[13]

$$V(R) = V_0\, e^{-aR} \tag{19}$$

with $a = 1.7\ a_0^{-1}$ and $V_0 = 250$ eV. The quasistatic detuning is then given approximately by (for simplicity of notation, $\hbar$ is set to 1 in this section so $\omega = \Delta E$)

$$\omega = V_f - V_i \simeq \frac{dV}{dR}\Delta R = -aV\,\Delta R\ ; \tag{20}$$

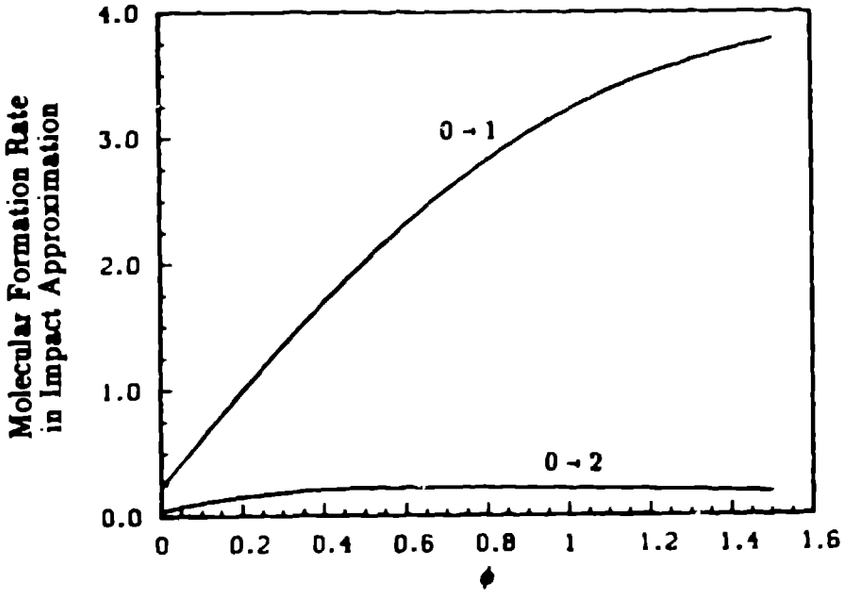

Fig. 5. Components of the molecular-formation rate (in arbitrary units) due to the 0→1 and 0→2 below-threshold resonances, *calculated using the impact-approximation width.*

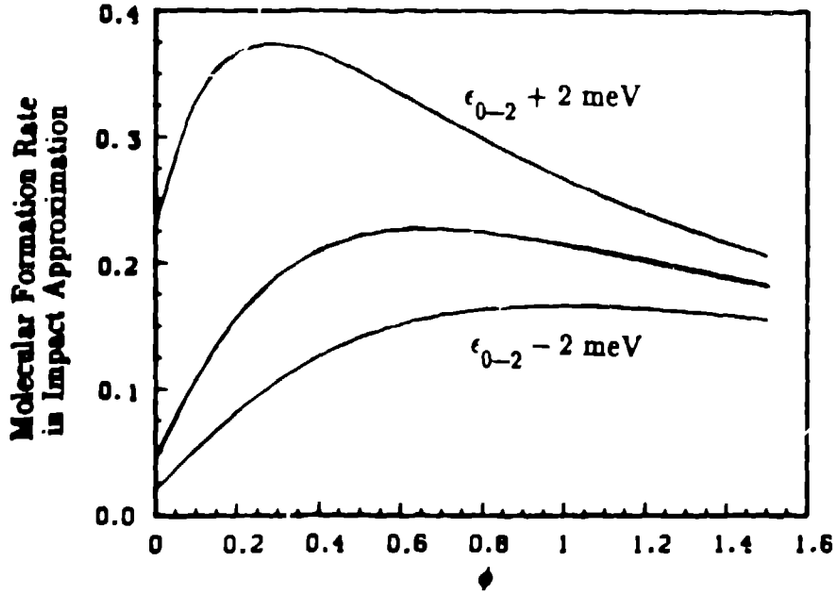

Fig. 6. Effect of raising or lowering the 0→2 resonance energy by 2 meV.

furthermore

$$\frac{d\omega}{dR} \simeq a^2 \, V \, \Delta R \ . \tag{21}$$

Using Eqs. (18)–(21) in Eq. (6), we obtain

$$I_{qs}(\omega) = \frac{C}{\omega} \int_0^{\pi} \ln^2\left[\frac{V_o a \, \Delta R}{\omega}\right] \exp\left[-\frac{4\pi n}{3a^3} \ln^3\left[\frac{V_o a \, \Delta R}{\omega}\right] - \frac{\omega}{a \, \Delta R \, kT}\right] \tag{22}$$

where ΔR is a function of θ and C is a density-dependent normalization constant. If ΔR is approximated by its average value, $\sim 0.09 \ a_0$, I_{qs} can be written as a simple analytic formula,

$$I_{qs}(\omega) \simeq \frac{C}{\omega} \ln^2\left[\frac{4\times 10^4}{\omega}\right] \exp\left[-0.0054 \ln^3\left[\frac{4\times 10^4}{\omega}\right] - 74 \, \frac{\omega}{T}\right] \tag{23}$$

for ω in meV and T in °K.

Figures 7 and 8 show the quasistatic line shape, evaluated by Eq. (22), for temperatures of 20 and 300°K at liquid-hydrogen density. For comparison, the Lorentzian from the impact approximation is also shown. Obviously the Lorentzian has a much broader wing. In contrast to the impact approximation, which predicts dominance of the 0→1 resonance over the 0→2 resonance at low temperatures, the quasistatic approximation predicts just the opposite; i.e., in the quasistatic case, the effect of the large detuning of the 0→1 resonance outweighs its larger matrix element. The profile in the quasistatic approximation also has a much stronger temperature dependence than in the impact approximation. The impact width at 300°K is only about 50% greater than at 20°K, whereas the quasistatic width increases by an order of magnitude.

It should be emphasized that the present treatment can be expected to bear only a qualitative resemblance to an accurate calculation for $(dt\mu)dee$ in a D_2 bath. Important interactions, at least in the near wing, are certainly not binary at liquid-hydrogen density. The exponential-interaction potential may also be misleading; e.g., it leads to a one-sided line shape having a blue wing only.

VI. CONCLUSIONS

Our understanding of the pressure broadening of the $(dt\mu)dee$ formation resonances is still far from complete. We are not yet able to make quantitative predictions, in part because of lack of a complete theory and in part because of inadequate knowledge of the binding energies. The line-broadening criteria due to target density and resonance detuning suggest that a quasistatic approximation will generally be more applicable to $(dt\mu)dee$ formation than will the impact approximation, the latter now having been accurately evaluated. This judgment is true in spite of the fact that $|\Delta E|$ is smaller than the impact width Γ. However, a rough quasistatic approximation seems to predict a stronger temperature dependence than has been experimentally observed. One possible explanation is that the resonance actually lies closer to threshold than predicted. Another possible explanation is that the quasistatic treatment needs

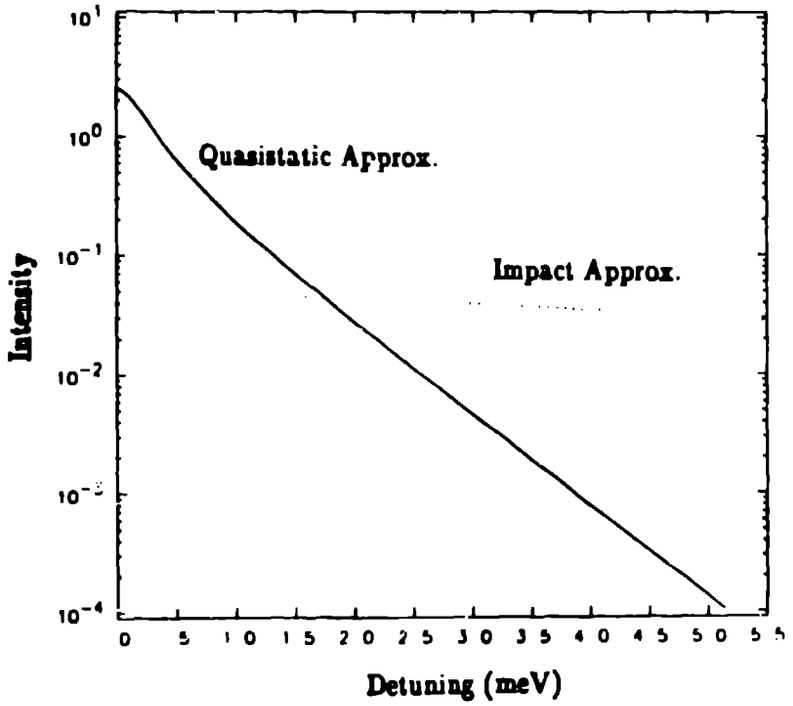

Fig. 7. Comparison of a crude quasistatic-approximation line shape (solid curve) with the impact-approximation line shape (dotted curve) at 20° K and LHD.

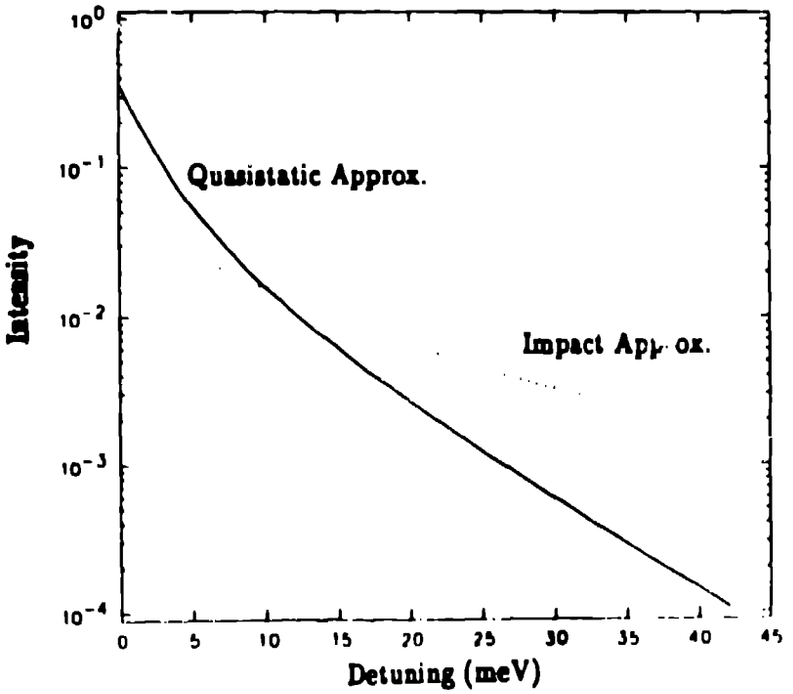

Fig. 8. Comparison of a crude quasistatic-approximation line shape (solid curve) with the impact-approximation line shape (dotted curve) at 300° K and LHD.

to be generalized to take into account target recoil.

ACKNOWLEDGMENT

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Figure Captions

Fig. 1. Schematic of $[(dt\mu)dee]^*$ formation resonances, $J_i \rightarrow J_f$, with heights roughly indicative of the size of the matrix element and abundance of the initial state in a low-temperature target.

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