

In the recoil energy spectrum of the $^{119m}\text{Te} \xrightarrow{\text{EC}} ^{119}\text{Sb}$ decay the lower part thereof is estimated to be small enough to retain the decaying atom in the original lattice site.^{2,3} This has been experimentally verified for the sources Sn ^{119m}Te and $\text{Sb}_2^{119m}\text{Te}_3$, in which the majority of ^{119}Sb was found in the Te site.² Therefore, an appreciable amount of ^{119}Sb should have been found remaining in Sn site of the $\text{SnSb}(^{119m}\text{Te})$ sample, if a part of ^{119m}Te had been distributed in the Sn site on crystallization. Accordingly, it is concluded from the exclusive distribution of ^{119}Sb in Sb site of the sample that no ^{119m}Te was stabilized in the Sn site on crystallization of the SnSb melt.

The observations above on the distribution of ^{119}Sb in SnTe and ^{119m}Te in SnSb can be interpreted in a unified manner considering the electronegativity of the three elements involved. Dilute Sb atoms can take part in the formation of SnTe lattice from a melt as both cationic and anionic point defects occupying the Sn and Te sites, supplying preferentially the deficient constituent in case of nonstoichiometric samples, because they have an

electronegativity value lying between those of Sn and Te. On the other hand, Te is exclusively incorporated into the anionic site of SnSb , since it is even more electronegative than the anionic component of the matrix.

The present work shows that Mössbauer emission spectroscopy is a useful method for the site-distribution study of dilute atoms in binary compounds. It would be interesting to apply this technique to other systems in order to study how electronegativity competes with other factors in determining the distribution of dilute impurity atoms in solids.

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Resonance state lifetimes from stabilization graphs

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The stabilization method (SM) pioneered by Taylor and co-workers¹ has proven to be a valuable tool for estimating the energies of long-lived metastable states of electron-atom, electron-molecule, and atom-diatom complexes. In implementing the SM one searches for eigenvalues arising from a matrix representation of the relevant Hamiltonian H which are "stable" as the basis set used to construct H is varied.

To obtain lifetimes of metastable states, one can choose from among a variety of techniques²⁻⁷ (e.g., phase shift analysis, Feshbach projection "golden rule" formulas, Siegert methods, and complex coordinate scaling methods), many of which use the stabilized *eigenvector* as starting information. Here we demonstrate that one can obtain an *estimate* of the desired lifetime directly from the stabilization graph in a manner which makes a close connection with the complex coordinate rotation method (CRM) for which a satisfactory mathematical basis exists.

The starting point of our development is the observation that both the stable eigenvalue (E_s) and the eigenvalue(s) (E_c) which come from above and cross E_s (see Fig. 1 and Refs. 9-11 and 13) vary in a nearly linear manner (with α) near their avoided crossing points. This observation leads us to propose that the two eigenvalues arising in each such avoided crossing can be

thought of as arising from two "uncoupled" states having energies $\epsilon_s(\alpha) = \epsilon + S_s(\alpha - \alpha_c)$ and $\epsilon_c(\alpha) = \epsilon + S_c(\alpha - \alpha_c)$, where S_s and S_c are the slopes of the linear parts of the stable and "continuum" eigenvalues, respectively. α_c is the value of α at which these two straight lines would intersect, and ϵ is their common value at $\alpha = \alpha_c$. This modeling of ϵ_s and ϵ_c is simply based upon the *observa-*

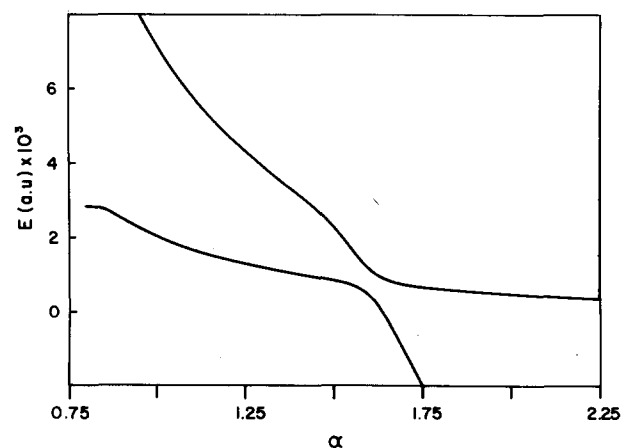


FIG. 1. Stabilization graph for the 2π shape resonance state of LiH^- (Ref. 9).

TABLE I. Stabilization graphs.

System/resonance type	α_c	ϵ (eV)	$ S_r / S_c $	V (eV)	Γ [Eq. (4)] (eV)	Other results (eV)
$H^-(^1S)$, Feshbach	1.33	9.52	0.018–0.035	0.27	0.14–0.20	$E = 9.56 - \frac{1}{2}i 0.048^a$ $E = 9.56 - \frac{1}{2}i 0.057^b$
$LiH^-(^2\pi)$, shape	1.6	0.02	0.10	9.6×10^{-3}	0.012	$E = 0.02 - \frac{1}{2}i 8.4 \times 10^{-3}^c$
A + BC $J=0$, Feshbach	0.96	128 cm^{-1}	0.14	3.25 cm^{-1}	4.86 cm^{-1}	$E = (114 - \frac{1}{2}i 2.1) \text{ cm}^{-1}^d$
$He(^1S)$, Feshbach	1.92	21.2	9.0×10^{-4}	0.87	0.10	$E = 21.2 - \frac{1}{2}i 0.13^e$

^aReference 8.^bReference 4.^cReference 9.^dReferences 10–12.^eReference 13.

tion that such approximately linear behavior occurs in most stabilization plots in the neighborhood of each avoided crossing. The matrix element $V(\alpha)$ which mixes the uncoupled states (ϵ_r, ϵ_c) to produce E_r and E_c is assumed to be localized near $\alpha = \alpha_c$ and to go to zero where E_r and E_c are linear.

The 2×2 secular problem whose elements are ϵ_r , ϵ_c , and V gives rise to two eigenvalues E_{\pm} :

$$E_{\pm} = \frac{1}{2}[\epsilon_r + \epsilon_c \pm \sqrt{(\epsilon_r - \epsilon_c)^2 + 4V^2}]. \quad (1)$$

To obtain $V(\alpha_c)$, we evaluate the difference between E_+ and E_- at $\alpha = \alpha_c$ and assume that this difference is equal to $E_r - E_c$ at α_c , which is easily read off the stabilization graph (SG) $E_r - E_c = 2|V(\alpha_c)|$.

If we assume (to make the simplest reasonable model) that $V(\alpha)$ is "peaked" at α_c and varies slowly about α_c , we can solve for the value of α at which $E_+ = E_-$:

$$\bar{\alpha} - \alpha_c = \pm \frac{2i|V(\alpha_c)|}{|S_r - S_c|} \equiv 2ig. \quad (2)$$

The relevance of this complex crossing point $\bar{\alpha}$ to the stable behavior demanded of resonant eigenvalues in the CR theory can be seen by investigating the behavior of E_+ and E_- in the neighborhood of $\bar{\alpha}$. Let us parametrize α in this region as $\alpha - \alpha_c = (\bar{\alpha} - \alpha_c)(1+z) = 2ig(1+z)$. The two eigenvalues E_{\pm} can be expressed as functions of z using Eqs. (1) and (2) and the fact that $(S_r - S_c)^2(\bar{\alpha} - \alpha_c)^2 + 4V^2 = 0$:

$$E_{\pm} = \epsilon - ig|S_r + S_c|(1+z) \pm |S_r - S_c|ig\sqrt{2z + z^2}. \quad (3)$$

Notice that real (imaginary) values of z correspond to imaginary (real) variations in α . It turns out that real variations in z affect most strongly the imaginary parts of E_{\pm} . Because we are interested in obtaining a reasonable estimate of the resonance *width*, it is on these real z variations that we focus our attention.

For positive x ($\text{Re } z$) the imaginary part of E_+ decreases uniformly with x and the imaginary part of E_- varies with x as $ig(|S_r - S_c|\sqrt{X^2 + 2X} - |S_r + S_c|X) - ig|S_r + S_c|$, which can pass through an extremum at $X = -1 + \sqrt{1 + [|S_r - S_c|^2 / (|S_r + S_c|^2 - |S_r - S_c|^2)]}$ if

$|S_r + S_c| > |S_r - S_c|$. For negative x , neither E_{\pm} has an extremum for real x .

We see therefore that *one* of the branches (E_{\pm}) may have an extremum depending both on whether $|S_r + S_c|$ or $|S_r - S_c|$ is larger and/or whether x is positive or negative. At a stable point dE/dx vanishes. For values of x beyond this stable point, the slope is no longer zero: it gains magnitude but *never* exceeds its large x limit of $-|S_r + S_c| + |S_r - S_c|$. Therefore, the *range of stability* of that branch (E_{\pm}) which has the extremum behavior is determined by $|S_r - S_c| - |S_r + S_c|$. This kind of behavior is what one refers to as a stable trajectory when applying the CR method to resonance states.

The value of E_{\pm} at the stable point discussed above can be obtained from Eqs. (3) and the definition of g :

$$E_{\pm} = \epsilon - 2i \frac{|V(\alpha_c)|}{|S_r - S_c|} \sqrt{|S_r||S_c|}. \quad (4)$$

This is our approximation to the CR theory's complex eigenenergy $E = \epsilon - i\Gamma/2$. We estimate the position of the resonance as ϵ (obtained as the crossing point of the two uncoupled curves) and the full width Γ as $4|V(\alpha_c)|\sqrt{|S_r||S_c|}/|S_c - S_r|$.

By using Eq. (4) to demonstrate how results are obtained in a "back of an envelope" fashion, we present in Table I an analysis of the SG's of Fig. 1 and those contained in Refs. 9, 10, and 13, together with the results of more accurate calculations on these same systems. These cases include shape and Feshbach resonances involving electronic and heavy particle states. The stabilization graphs were chosen to be representative rather than ideal (i. e., not all graphs have extremely linear regions of E_r and E_c , and on some of the graphs the estimation of S_r and S_c is difficult). The results obtained via Eq. (4) seem to lie within a factor of 4 of the presumably accurate values reported in Table I. Therefore, there is reason for optimism about the potential utility of Eq. (4) as a means of *estimating* the resonance lifetime. Clearly, when it is difficult to identify linear regions of the SG or to evaluate the slopes (S_r and S_c), its utility is to be questioned.

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Pulsed molecular beams: A lower limit on pulse duration for fully developed supersonic expansions

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Pulsed nozzle sources offer advantages in many kinds of molecular beam experiments.¹ Several designs utilizing pulse durations of 10 μ s to 2 ms are now available.^{2,3} However, it has remained unclear what pulse durations are required if a pulsed nozzle is to produce a supersonic beam comparably "cold" to that obtained from a continuous flow nozzle source.

The minimum time that the source valve is completely open is denoted by Δt_{\min} . This is a sum⁴ of three terms: Δt_1 , the "initiation" time required for the gas behind the nozzle to be accelerated to the (sonic) exit velocity; Δt_2 , the time required, assuming continuous flow conditions, to establish a buffer zone containing sufficient molecules to act as collision partners during the expansion; and Δt_3 , the time for which the pulse must be on to insure that, in the region over which the expansion takes place, the density in the buffer zone is not diminished by the velocity dispersion at the pulse edges. For valve open times longer than Δt_{\min} , the terminal beam temperatures are independent of pulse length.

Our calculation for Δt_1 is based on the fact⁵ that, in a continuous expansion, the acceleration from rest to the sonic velocity u_1 ⁶ takes place within one nozzle diameter D of the nozzle exit. Assuming a constant acceleration, we obtain

$$\Delta t_1 = 2D/u_1 = (2D/u_\infty)[(\gamma + 1)/(\gamma - 1)]^{1/2}, \quad (1)$$

where γ is the specific heat ratio C_p/C_v , and u_∞ (the zero enthalpy limit of the flow velocity) is

$$u_\infty = [\gamma/(\gamma - 1)]^{1/2} [2kT_0/m]^{1/2}, \quad (2)$$

where T_0 is the stagnation temperature.

Our calculations for Δt_2 and Δt_3 employ the "sudden freeze" model,⁶⁻⁸ in which the local temperature T , density n , and flow velocity u are assumed to be in thermodynamic equilibrium until some "freezing distance" from the nozzle, denoted x_F . At x_F , free molecular flow commences, and collisions beyond x_F are so infrequent as to leave $T(x)$ and $u(x)$ essentially unchanged from their values at x_F , denoted T_F and u_F . The number of collisions that occur in the remaining expansion beyond x_F is denoted N_F . This is an adjustable parameter, but we expect $N_F \approx 1$, e.g., a fit to measurements of Anderson and Fenn⁹ gives $N_F = 0.8$ for an argon expansion. The freezing distance is given by

$$x_F = x_{\text{ref}} [(8(\gamma - 1)/\pi\gamma)^{1/2} (x_{\text{ref}}/D) (\pi\sigma^2 P_0 D) (kT_0 N_F)^{-1}]^{1/\gamma}, \quad (3)$$

with P_0 the stagnation pressure, D the nozzle diameter, $\pi\sigma^2$ the hard sphere collision cross section, and

$$x_{\text{ref}} = D [(\gamma - 1)A^2/2]^{-1/2(\gamma-1)}. \quad (4)$$

Here $A(\gamma)$ is a function first defined by Ashkenas and Sherman⁵ and tabulated elsewhere^{8,10}; $A(5/3) = 3.26$ and $A(7/5) = 3.65$.

For expansions involving molecules with internal degrees of freedom, N_F^{rot} and N_F^{vb} can be similarly defined, e.g., N_F^{vb} is the number of collisions remaining in the expansion after the vibrational temperature T^{vb}