Nonvanishing $J=1\leftrightarrow 0$ equal-frequency two-photon decay: $E1M2$ decay of the He-like $2\,^3S_1$ state

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(Received 4 February 2004; published 3 June 2004)

The selection rule which forbids $J=1\leftrightarrow 0$ two-photon decay when the energies of the two photons are equal is shown to apply only when the multipolarities of the two photons are the same (i.e., $2E1$, $2M1$, etc.) and does not generally apply to mixed multipolarity amplitudes such as $E1M2$. A calculation of the two-photon decay rate for He-like ions in the $2\,^3S_1$ state including both $2E1$ and $E1M2$ two-photon decay amplitudes is presented. It shows a significant contribution for the case of equal-energy photons. The ratio of the $E1M2$ contribution to the $2E1$ contribution is found to be surprisingly large. For example, it is 0.27 for two-photon decay of the $2\,^3S_1$state in He-like uranium.

DOI: 10.1103/PhysRevA.69.062502 PACS number(s): 31.10.+z, 32.80.Wr, 32.70.Cs

I. INTRODUCTION

The selection rules for two-photon transitions in atoms have been discussed by a number of authors [1–7]. A summary for the case when both photons have the same energy has been given by Grynberg and Cagnac [6] and the more general multifrequency case has been discussed by Bonin and McIlrath [5]. One important selection rule forbids $J=1\leftrightarrow 0$ two-photon transitions when the photons have the same energy. (J is the electronic angular momentum.) DeMille *et al.* [8] emphasize that this rule is a consequence of the exchange symmetry of photons required by Bose-Einstein statistics and they tested it experimentally in a search for exchange-antisymmetric two-photon states. It is important to point out, however, a limitation in all of the discussions of this selection rule, that only the lowest order amplitude involving emission of two electric dipole photons ($2E1$) has been considered.

A stronger selection rule applies to the disintegration of $^3S_1$ positronium into two (equal-energy) photons. It can be shown using fundamental symmetry arguments that this process is forbidden to all orders, in fact any spin-1 particle (either vector or pseudovector) is forbidden to disintegrate into two photons [9–11]. The proof of this rule depends on the fact that, following disintegration, there are two equal-energy, counter-propagating photons in the center of mass [9,10]. This condition does not generally apply in atomic two-photon decay where the final state consists of three bodies: two photons plus a recoiling atom. So we normally cannot apply the stronger selection rule to atomic two-photon decay.

In this paper, we consider whether the selection rule which forbids $J=1\leftrightarrow 0$, equal frequency, two-photon decay in atoms applies for multipolarities beyond $2E1$. We use the standard formulas for atomic two-photon decay and find that the rule does apply for all amplitudes in which both photons have the same multipolarity (i.e., $2E1$, $2M1$, $2E2$, etc.), but that it does not apply to mixed multipolarity amplitudes such as $E1M2$, $E2M1$. We discuss the derivation of an extended selection rule in Sec. II, and in Sec. III we explore the significance of the result by making an approximate calculation of the leading order mixed multipolarity amplitude ($E1M2$) for the two-photon decay rate of the $2\,^3S_1$ level in heavy He-like systems. This is a good test case because extensive calculations of the $2E1$ decay rate for this system have been done [4].

II. DERIVATION OF THE SELECTION RULE

The relativistic formula for the two-photon decay rate (in atomic units) given by Goldman and Drake [12] is

$$\frac{dw}{d\omega_1}=\frac{\omega_1\omega_2}{(2\pi)^3c^2}\left|\sum_n\frac{\langle f|\tilde{A}_2^*|n\rangle\langle n|\tilde{A}_1^*|i\rangle}{E_n-E_i+\omega_1}+\frac{\langle f|\tilde{A}_1^*|n\rangle\langle n|\tilde{A}_2^*|i\rangle}{E_n-E_i+\omega_2}\right|^2 d\Omega_1 d\Omega_2. \quad (1)$$

The operator for photon j can be expanded in partial waves (see Ref. [12]):

$$\tilde{A}_j^*=\sum_{\lambda,L,M}[\hat{e}_j\cdot\vec{Y}_{LM}^{(\lambda)}(\hat{k}_j)]\tilde{a}_{LM}^{(\lambda)}(r)^*. \quad (2)$$

The symbols $\hat{e}_j$ and $\hat{k}_j$ refer to the polarization and momentum vectors of the photon, and the $\vec{Y}_{LM}^{(\lambda)}(\hat{k}_j)$ are related to the vector spherical harmonics [12]. The $\tilde{a}_{LM}^{(\lambda)}(r)^*$ are spherical tensor operators of rank L, and λ indicates the type of multipole; $\lambda=1$ for electric multipoles and $\lambda=0$ for magnetic multipoles.

For simplicity, we assume the atom has a spinless nucleus but the extension to the general case of nonzero nuclear spin is straightforward [6]. We consider an initial state with electronic angular momentum J_i and a final state with $J_f=0$. Substituting the photon operators from Eq. (2) into Eq. (1), applying the Wigner-Eckart theorem, and simplifying using the properties of the $3j$ symbols [13], the nth term in the sum in Eq. (1) becomes

$$\sum_{LL'MM'\lambda\lambda'} [\hat{e}_1 \cdot \vec{Y}^{(\lambda')}_{L'M'}(\hat{k}_1)][\hat{e}_2 \cdot \vec{Y}^{(\lambda)}_{LM}(\hat{k}_2)]\begin{pmatrix} L & L' & J_i \\ M & M' & M_i \end{pmatrix} \times \left[\frac{\langle \gamma_f 0 \| \widetilde{a}^{(\lambda)*}_L \| \gamma_n J_n \rangle \langle \gamma_n J_n \| \widetilde{a}^{(\lambda')*}_{L'} \| \gamma_i J_i \rangle}{\sqrt{2L+1}(E_n - E_i + \omega_1)} + (-1)^{L+L'+J_i} \frac{\langle \gamma_f 0 \| \widetilde{a}^{(\lambda')*}_{L'} \| \gamma_n J_n \rangle \langle \gamma_n J_n \| \widetilde{a}^{(\lambda)*}_L \| \gamma_i J_i \rangle}{\sqrt{2L'+1}(E_n - E_i + \omega_2)} \right]. \quad (3)$$

The $\gamma_{i,f,n}$ designate all the other quantum numbers of the states. The phase factor $(-1)^{L+L'+J_i}$ ensures the proper exchange symmetry required by Bose-Einstein statistics. All the dependence on the photon polarizations and angular correlations is contained in the factor

$$[\hat{e}_1 \cdot \vec{Y}^{(\lambda')}_{L'M'}(\hat{k}_1)][\hat{e}_2 \cdot \vec{Y}^{(\lambda)}_{LM}(\hat{k}_2)].$$

We are interested in the total decay rate so we will integrate over all photon directions and polarizations.

For the degenerate frequency case ($\omega_1 = \omega_2$), if the multipolarities of the two photons are the same (i.e., $2E1, 2M1, 2E2$, etc.) and J_i is odd, then the bracketed sum in Eq. (3) vanishes since the two terms have the same magnitude and the phase factor is negative. In particular, for a $2E1$ amplitude each term in the summation indicated in Eq. (3) vanishes for $J_i = 1$, which demonstrates the selection rule, forbidding $J = 1 \leftrightarrow 0$ two-photon transitions for equal-energy photons. In the same way one can show that the rule also holds for $J = 0 \rightarrow 1$ transitions.

For mixed multipolarity two-photon transitions ($E1M2, E2M1$, etc.), the magnitudes of the numerators of the two terms in Eq. (3) are not generally equal so they do not exactly cancel when the frequencies are degenerate even for the cases where the phase factor $(-1)^{L+L'+J_i}$ is negative. Also, since we are concerned with the two-photon decay rate that requires integration over the angles of emission and polarizations of the photons, there can be no cancellation due to interferences between the different terms in the summation of Eq. (3). This is because cross terms involving different multipoles for the same photon vanish in the integration over photon directions and polarizations [12]. So the rule forbidding equal-frequency $J = 1 \leftrightarrow 0$ two-photon decay applies only when all mixed multipolarity amplitudes are negligible.

III. TWO PHOTON DECAY OF THE He-LIKE $2\,^3S_1$ STATE

As an example of a mixed multipolarity transition which illustrates a non-negligible equal frequency $J = 0 \leftrightarrow 1$ two-photon decay, we present a calculation of the two-photon decay rate of the He-like $2\,^3S_1$ state including both $2E1$ and $E1M2$ amplitudes. At first glance one might expect the $E1M2$ amplitude to be uninteresting since it is usually a very good approximation to neglect all multipoles beyond $2E1$ in calculating two-photon decay rates between atomic states of the same parity. For example, Goldman and Drake [12] found that the ratio of the $E1M2$ contribution to the $2E1$ contribution to the two-photon decay of a H-like $(2\,^2S_{1/2})$ ion of atomic number Z is $3.08 \times 10^{-11} Z^4$. Even for uranium this ratio is only 2.2×10^{-3}. On the other hand, the $2E1$ amplitude for decay of the $2\,^3S_1$ level in He-like ions is suppressed because the direct and exchange terms add incoherently. There is no similar suppression for the $E1M2$ amplitude in the decay of the $2\,^3S_1$ level since here the direct and exchange terms add coherently. Thus the $E1M2$ amplitude should be relatively more important for the two-photon decay of the He-like $2\,^3S_1$ level than it is for decay of the H-like $2\,^2S_{1/2}$ level. Furthermore, the higher order two-photon amplitudes increase more rapidly with Z than $2E1$ does, suggesting that it is particularly important to consider the mixed multipolarity terms at high Z.

The He-like $2\,^3S_1$ state decays primarily to the $1\,^1S_0$ ground state by single photon $M1$ emission but it can also decay by two-photon emission. The two-photon decay branch has been analyzed in the nonrelativistic approximation by Bely and Faucher [2,3] and by Drake *et al.* [1]. Derevianko and Johnson [4] have done an accurate relativistic calculation which covers $Z=2$ to $Z=100$. In all of these calculations only the lowest order $2E1$ amplitude was retained.

In carrying out a new calculation of the two-photon decay rate of the $2\,^3S_1$ state, the aim is to assess the importance of the $E1M2$ amplitude relative to the $2E1$ amplitude. Since high accuracy is not required, we neglect electron-electron interactions of order $1/Z$ but take relativistic effects fully into account so that the results will have meaning at high Z. Our approach is similar to that used by Drake [14] to calculate the $E1M1$ decay rate of the $2\,^3P_0$ level in He-like ions.

We begin with He-like wave functions that are jj-coupled products of Dirac single particle wave functions:

$$\Psi(\vec{r}_1, \vec{r}_2) = \frac{1}{\sqrt{2}} \left\{ \sum_{\mu,m} \Psi_1(1s_{1/2}, \mu) \Psi_2(nljm) \left\langle \frac{1}{2} \mu jm \middle| JM \right\rangle - \text{exchange}. \right. \quad (4)$$

We substitute these wave functions into Eq. (1) with the initial state $1s2s\,^3S_1$, and the final state $1s^2\,^1S_0$. The only states that survive the sum over n are (in jj coupling): $(1s_{1/2}, np_{1/2} J=1)$, $(1s_{1/2}, np_{3/2}, J=1)$, and $(1s_{1/2}, np_{3/2}, J=2)$. We use the formalism of Goldman and Drake [12] to reduce the two-electron matrix elements in Eq. (1) to one-electron radial integrals, integrating over photon directions and polarizations and averaging over the magnetic quantum numbers of the initial state. This gives a differential decay rate of

$$\frac{dw}{d\omega_1} = \frac{\omega_1 \omega_2}{\pi c^2} \frac{2^3}{3^6} \{ |Q_{2E1}(\omega_1, \omega_2) - Q_{2E1}(\omega_2, \omega_1)|^2 + |Q_{E1M2}(\omega_1, \omega_2)|^2 + |Q_{E1M2}(\omega_2, \omega_1)|^2 \}. \quad (5)$$

Here,

$$Q_{2E1}(\omega_1,\omega_2)=\sum_n\left\{\frac{M^{(1,1)}_{1s_{1/2},np_{1/2}}(\omega_1)M^{(1,1)}_{np_{1/2},2s_{1/2}}(\omega_2)}{E(np_{1/2},1)-E(2s_{1/2},1)+\omega_2}-\frac{M^{(1,1)}_{1s_{1/2},np_{3/2}}(\omega_1)M^{(1,1)}_{np_{3/2},2s_{1/2}}(\omega_2)}{E(np_{3/2},1)-E(2s_{1/2},1)+\omega_2}\right\}, \quad (6)$$

and

$$Q_{E1M2}(\omega_1,\omega_2)=\frac{3\sqrt{3}}{5}\sum_n\left\{\frac{M^{(1,1)}_{1s_{1/2},np_{3/2}}(\omega_1)M^{(0,2)}_{np_{3/2}2s_{1/2}}(\omega_2)}{E(np_{3/2},1)-E(2s_{1/2},1)+\omega_2}-\frac{M^{(0,2)}_{1s_{1/2},np_{3/2}}(\omega_2)M^{(1,1)}_{np_{3/2}2s_{1/2}}(\omega_1)}{E(np_{3/2},2)-E(2s_{1/2},1)+\omega_1}\right\}. \quad (7)$$

The radial integrals $M^{(\lambda,L)}_{\alpha\beta}(\omega)$ are given by Ref. [12]:

$$M^{(1,1)}_{\alpha,\beta}=\frac{1}{\sqrt{2}}[(\kappa_\alpha-\kappa_\beta)I^+_2+2I^-_2]-\sqrt{2}[(\kappa_\alpha-\kappa_\beta)I^+_0-I^-_0]+G[3J_1+(\kappa_\alpha-\kappa_\beta)(I^+_2+I^+_0)-I^-_0+2I^-_2], \quad (8)$$

$$M^{(0,2)}_{\alpha,\beta}=\frac{5}{\sqrt{6}}(\kappa_\alpha+\kappa_\beta)I^+_2, \quad (9)$$

where

$$I^\pm_L=\int_0^\infty(g_\alpha f_\beta\pm f_\alpha g_\beta)j_L(\omega r/c)dr, \quad (10)$$

$$J_L=\int_0^\infty(g_\alpha g_\beta+f_\alpha f_\beta)j_L(\omega r/c)dr. \quad (11)$$

Here, $M^{(1,1)}_{\alpha,\beta}$ and $M^{(0,2)}_{\alpha,\beta}$ are the electric dipole and magnetic quadrupole integrals, $j_L(kr)$ is a spherical Bessel function, g_α and f_α are the large and small components of the Dirac H-like wavefunctions, and κ_α is the usual Dirac quantum number for these states. The parameter G is an arbitrary gauge parameter. The velocity gauge corresponds to $G=0$ and the length gauge corresponds to $G=\sqrt{2}$.

The total decay rate is obtained by integrating Eq. (5) over ω_1,

$$w^{2\gamma}=\frac{1}{2}\int_0^{\omega_0}\frac{dw}{d\omega_1}d\omega_1, \quad (12)$$

where ω_0 is the transition frequency. The factor of 1/2 is needed because photon 1 is counted twice in the interval $\{0,\omega_0\}$.

The sum over n in Eqs. (6) and (7) includes an infinite sum over all discrete states and integrals over all positive-energy and negative-energy continuum states. We use the finite basis set (FBS) method of Drake and Goldman [12,15], to reduce the infinite sum and the integrals to a finite sum over discrete variational solutions to the Dirac equation. Convergence of both the $2E1$ and $E1M2$ parts of the total decay rate to better 0.01% is obtained with 16 basis states (eight positive energy and eight negative energy). For the energies of the $n=1$ and $n=2$ states, we substitute accurate values calculated by Drake [16] in place of the FBS energies. This makes the calculations gauge dependent. Calculations were done in both length and velocity gauges to provide some indication of the accuracy of the results. In Table I we compare the results of the $2E1$ and $E1M2$ calculations in both length and velocity gauge. For the $2E1$ amplitude, the difference is about 15% while for $E1M2$ the disagreement between the gauges becomes smaller as Z increases, being less than 4% at $Z=80$.

Table II presents a summary of the results of our calculation for a number of values of Z between 40 and 100. To get better accuracy for the corrected total two-photon decay rates, we use the accurate values of Derevianko and Johnson for the $2E1$ part and our calculations in the length gauge for

TABLE I. Comparison of results of the present calculation for length (L) and velocity (V) gauges for the $2E1$ and $E1M2$ parts of the decay rates (s^{-1}) of the $2\,^3S_1$ state of heliumlike ions. Numbers in square brackets indicate the power of ten by which to multiply.

Z	$2E1(L)$	$2E1(V)$	$E1M2(L)$	$E1M2(V)$
40	8.952[6]	10.29[6]	1.311[6]	1.405[6]
60	3.925[8]	4.766[8]	7.822[7]	8.197[7]
80	5.495[9]	6.506[9]	1.353[9]	1.405[9]
100	3.950[10]	4.617[10]	1.194[10]	1.236[10]

TABLE II. Two-photon $A_{2\gamma}$ and $M1$ A_{M1} decay rates (s^{-1}) for decay of the $2\,^3S_1$ state in He-like ions. The two-photon decay rate is broken down into contributions from the $2E1$ and $E1M2$ multipoles. Numbers in square brackets indicate the power of ten by which to multiply.

Z	$A_{2\gamma}(2E1)$[a]	$A_{2\gamma}(E1M2)$[b]	$A_{2\gamma}$	A_{M1}[c]
40	7.69[6]	1.31[6]	9.00[6]	1.719[10]
45	2.46[7]	4.36[6]	2.90[7]	5.792[10]
50	6.88[7]	1.26[7]	8.14[7]	1.726[11]
55	1.72[8]	3.28[7]	2.05[8]	4.658[11]
60	3.93[8]	7.82[7]	4.71[8]	1.161[12]
65	8.34[8]	1.74[8]	1.01[9]	2.706[12]
70	1.66[9]	3.62[8]	2.02[9]	5.968[12]
75	3.14[9]	7.07[8]	3.85[9]	1.256[13]
80	5.65[9]	1.35[9]	7.00[9]	2.540[13]
85	9.78[9]	2.46[9]	1.22[10]	4.968[13]
90	1.63[10]	4.30[9]	2.06[10]	9.439[13]
92	1.98[10]	5.32[9]	2.51[10]	1.212[14]
95	2.64[10]	7.17[9]	3.36[10]	1.751[14]
100	4.15[10]	1.19[10]	5.34[10]	3.181[14]

[a]Derevianko and Johnson [4].
[b]This work using length gauge ($G=\sqrt{2}$).
[c]Johnson, Plante, and Sapirstein [17].

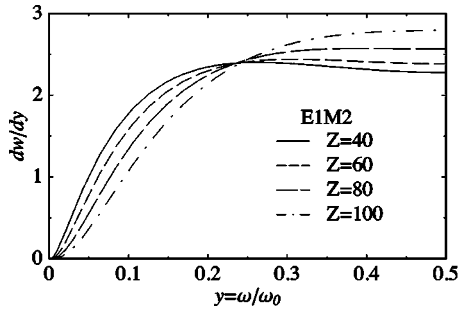

FIG. 1. Lower half of the continuum distribution for the $E1M2$ part of the total decay rate as a function of the fractional energy $y = \omega/\omega_0$. The curves are normalized to area 2 over the full interval $y=0$ to $y=1$.

the $E1M2$ part. The $E1M2$ contribution is a significant fraction of the rate throughout this range of Z. The ratio of the $E1M2$ amplitude to the $2E1$ amplitude increases from 0.17 at $Z=40$ to 0.29 at $Z=100$, so the additional amplitude is important in the medium to high Z regime. For completeness we also include the $M1$ decay rate [17] since this is the dominant decay mechanism for the $2\,^3S_1$ state in He-like ions.

In Fig. 1 we give the differential decay rate dw/dy as a function of $y=\omega/\omega_0$ for the $E1M2$ part for several values of Z. At the highest Z, the curves show a broad maximum at half the transition energy ($y=0.5$) falling to zero at either endpoint. The distributions at lower Z are similar except that there is a slight dip at the center and the maxima shift to either side of the center. The main feature of interest here is that these curves do not vanish at the point $y=0.5$ where the energies of the two-photons are equal. Figure 2 shows the spectral distributions of the combined $2E1$ and $E1M2$ contributions to the decay rate for several values of Z. These show peaks at low energy that are sharpest at low Z and increase in width as Z increases. The curves have minima at $y=0.5$ but do not go to zero there due to the contribution from the $E1M2$ part. Figure 3 shows the spectral distributions of the total decay rate and the contributions to the spectral distribution from $2E1$ and $E1M2$ multipoles for $Z=40$ (upper part) and $Z=92$ (lower part). In both cases the $2E1$ component vanishes at $y=0.5$. The region from $y=0.3$ to $y=0.5$ is dominated by the $E1M2$ contribution.

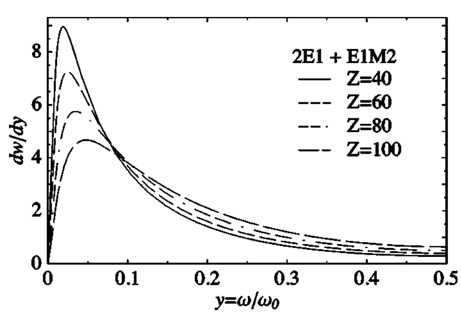

FIG. 2. Low-energy part of the He-like $2\,^3S_1$ two-photon continuum distribution as a function of the fractional energy $y=\omega/\omega_0$ normalized to area (=2 photons) over $y=0$ to $y=1$. (Present calculation in length gauge.)

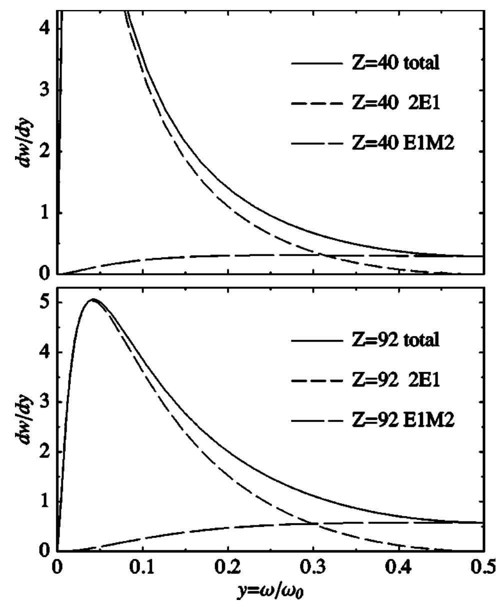

FIG. 3. Lower energy part of the spectral distributions of the $2E1$ and $E1M2$ contributions to the He-like $2\,^3S_1$ two-photon decay rate as a function of the fractional energy $y=\omega/\omega_0$. (Present calculation in length gauge.) The sum of the two contributions (total) is also given. The upper plot is for $Z=40$ and lower plot is for $Z=92$. All curves are normalized to area (=2 for the "total" curves over $y=0 \rightarrow 1$).

IV. CONCLUSION

We have shown that for two-photon decay of an atom with a change in electronic angular momentum $J_i=J \rightarrow J_f=0$, the decay rate vanishes for degenerate frequencies ($\omega_1=\omega_2$) if J is odd and the multipolarities of both photons are the same. This is in agreement with the well-known rule that the $2E1$ decay rate for $J=1 \leftrightarrow 0$ transitions vanishes when the energies of the two photons are equal. There is no similar rule that applies to mixed multipolarity transitions ($E1M2, E2M1$, etc.) and we find that two-photon decay rates can, in general, be nonvanishing when the frequencies of the two photons are equal.

To illustrate the significance of these results, we have calculated the decay rate of the $2\,^3S_1$ level in He-like ions including both the $2E1$ amplitude and the leading mixed multipolarity amplitude $E1M2$. The $E1M2$ amplitude dominates the spectral distribution for the $2\,^3S_1$ decay over a broad region in the center of the distribution. The $E1M2$ amplitude makes a surprisingly large contribution to the two-photon decay rate for the $2\,^3S_1$ state of He-like ions contributing 15% to the decay rate at $Z=40$ and 22% at $Z=92$.

The two-photon decay branch of the $2\,^3S_1$ state has not been observed to date, but the results of the calculation of the $E1M2$ amplitude provide some motivation for an experimental effort to study this decay branch. The relatively large contribution of the $E1M2$ amplitude and its separation from the $2E1$ contribution in the two-photon continuum spectrum provide an opportunity for a study in atomic physics of a two-photon amplitude beyond $2E1$. Also, the spectral distribution of the $E1M2$ part makes observation of the $2\,^3S_1$ two-photon decay more practical. This is because two-photon decay experiments are most sensitive at the center of the continuum distribution [18] and this is where the $E1M2$ amplitude contributes most strongly.

ACKNOWLEDGMENTS

The author has benefited from discussions with Murray Peshkin, Elliot Kanter, and Mitio Inokuti. This work was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences.

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