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Machine-learning predictions of positron binding to molecules

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Machine-learning techniques are used to check the theoretical and experimental predictions of positron binding to general molecules. The bound or unbound character of previous calculations for polar molecules are mostly confirmed. Binding for so far unexplored polar molecules is predicted. For apolar molecules, a formula for the binding energy in terms of isotropic polarizability and ionization potential is obtained, leading to unprecedented agreement with experiments as well as prediction of previously unidentified bound systems. The role of the ionization potential is suggested as a consequence of enhanced formation of virtual positronium at short distances.

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I. INTRODUCTION

The interaction of a slow positron (e^+) with atoms and molecules is important to many fields of science [1] and has been the subject of intensive experimental and theoretical study in recent decades [2]. Particularly interesting is the possibility of a positron forming bound states with such targets, which is fundamental to the understanding of enhanced annihilation rates observed in low-energy positron scattering experiments with gases [2]. In this process, a positron transfers part of its energy to molecular vibrations and is captured to form a bonded complex. The consequent resonant annihilation appears to be the only experimental confirmation of the existence of such bound states. Theoretical explanations of these phenomena assume empirically the existence of these states [2,3], despite the appearance of alternative explanations [4,5]. Other predictions come from calculations for atoms or polar molecular targets.

There is evident interest in understanding the ability of a molecule to bind a positron from its electronic properties. Molecular dipole moment (μ) and polarizability (α) are two obvious related properties, although insufficient as will be seen below. These quantities are also not independent, which makes this understanding even more difficult. In the present work, a method that addresses these difficulties is presented.

It was predicted long ago that a rigid dipole with μ larger than 1.625 D (Debye) would bind a unit charge [6]. Polar molecules are not rigid dipoles, however. In fact, the electronic effects of the interaction with an e^+ [7] on molecular dipole moments cannot be neglected. It was also shown that critical dipole moments are increased by rotations [8].

Binding to polar molecules is also influenced by the molecular polarizability. The existence of an almost critical dipole moment for positron binding, namely $\mu \simeq 1.85$ D, appears to be a consequence of the dependence of these two quantities. In fact, increasing dipole moments of small molecules is normally followed by decreasing polarizability. *Ab initio* calculations for molecules with larger μ are convincing in the prediction of binding, in spite of the still large differences from experimental binding energy (ϵ_b) values [9–22]. Despite the physics being rather complex, it appears that the localization of e^+ close to the negative pole of the target favors a description in which it occupies a localized orbital, hence helping calculations to yield reliable results. A model-potential approach for molecular targets was recently applied to e^+-HCN, with the predicted value of ϵ_b yet to be checked [23].

Due to the complexity of the electronic properties of larger atoms as well as to the larger binding energies involved, *ab initio* [24] or model potential [25] calculations for e^+-atom complexes present further obstacles. A recent survey shows that different calculations can disagree by as much as tenths of eVs [24]. Positron interactions with atoms can be considered a research field by itself, with particular experimental and theoretical approaches.

Apolar molecules are also difficult targets for calculations, which disagree among themselves as well as with experiments, mainly due to the positron delocalization. This is still an open problem so that attempts have been made to analyze these complexes using improved charge-polarizable target model potentials [26,27]. For these apolar systems (or those having small μ), the interaction is surely dominated by the molecular polarizability. A recent work proposed that an additive (over atoms) polarizability should be used instead of the isotropic one for alkanes, in order to achieve good agreement with experiment [27]. However, being a medium-to-large-distance interaction, polarization is not enough to fully explain binding. This is because, at short positron-target distances, electron-positron correlation plays an important role, yielding a tendency to form a positronium atom Ps (e^+e^- pair). If the positron energy is larger than ($E_i - 6.8$) eV, where E_i is the ionization potential of the target and 6.8 eV is the Ps ground-state energy, a Ps can be formed.

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Meanwhile, previous works suggest that assuming the contribution of a virtual Ps (formed by tunneling of an electron from the atom to the positron) is relevant for describing scattering [28], binding [29], and annihilation rates [30] of e^+ with atoms. Connected ideas involving a Ps component of the wave function of e^+-atom bound states are found in a review [31]. Previous studies recognize an independent role for E_i in positron binding to atoms [25,32]. It becomes arguable that the formation of the virtual Ps, the main effect of electron-positron correlation at short e^+-molecule distances, is connected to E_i, which should then play a role in e^+ binding, though not fully independent of α, even at low energy.

In Ref. [27], the inclusion a cutoff function in the potential is assumed to partially account to virtual Ps effects. An empirical approach like this, is hardly extendible to all kinds of apolar molecules, however. Furthermore, empirically cutting off the potential contributes little to understanding the phenomena involved since it has the role of regularizing the polarization potential, otherwise non-Hermitian [26] due to the well-known ill behavior at the origin of its $1/r^4$ dependence. A general approach based on physical properties of the target, which already accounts for short-range repulsive effects, should not depend on this artifact.

Fortunately, the availability of experimental values of ϵ_b with an accuracy of 10 meV for some families of molecules [33–35], allows the use of artificial intelligence techniques, such as machine learning (ML) [36], to accurately deal with known and previously unidentified complexes as well as to yield a deeper understanding of the problem, with no resort to *ad hoc* repulsive potentials. Instead, input data should be measurable properties of isolated molecules. Thus, inspired by approximate linear dependencies of ϵ_b on α and μ, suggested in Ref. [33], the inclusion of E_i together with α and μ is performed in the present work via a ML approach, as a property related to virtual Ps formation.

II. MACHINE-LEARNING METHOD

The sure independence screening and sparsifying operator (SISSO) ML routine [37,38] seems appropriate for the present purposes. In abstract, SISSO searches for dimensionality reduction of input data basis (the *training set*), e.g., physical properties of the systems under study (*features* in Refs. [37,38]) by testing huge numbers of analytical expressions (*descriptors*) involving these data. Then, SISSO searches for the simplest relations between the descriptors that yield a *classification* of the systems characterized by the input data. These techniques are capable of identifying quantities irrelevant to the problem, further optimizing the input data space. Among the input data, those belonging to a particular class (e.g., ϵ_b in our case) can be treated as a *subject* (*target* in Refs. [37,38]). SISSO yields as many descriptors as necessary for both classification or *regression*, the latter meaning to obtain an analytical expression for the subject in terms of the other input data, through a least-squares fitting of a linear combination of the descriptors. Here, classification is used to separate the complexes into bound and unbound categories, to check theoretical predictions, and to predict previously unidentified bound systems. Regression allows for checking the accuracy of previous calculations and to make predictions of ϵ_b for so far unknown cases. The remarkable advance allowed by the ML technique here is that the best descriptors are sought by the algorithm itself, among thousands of possibilities, not by a common method of adjusting points to known functions.

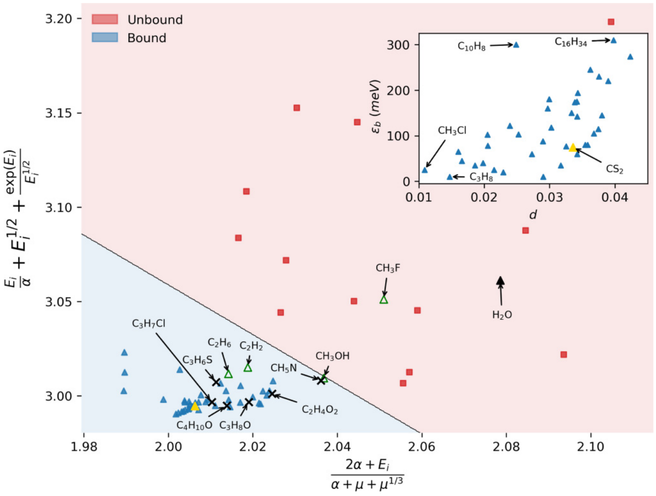

FIG. 1. Bound-unbound separation of molecular positron complexes. Blue triangles represent molecules of the training set. Green and yellow triangles are checking cases (see text). The molecules labeled with × have their bound character predicted here. The inset shows ϵ_b versus the distance d of each point to the separation line. Molecular properties data are taken from the experimental sources [33–35], except the predictions for which data are taken from NIST [39]. Data for the descriptors are in a.u.

III. RESULTS

A. Classification

For bound-unbound classification, the bound systems included in the training set are 37 polar and apolar molecules among the experimental data in Refs. [33,35]. Exceptions are isotopologues; complexes with ϵ_b lower than the experimental error of 10 meV; $C_4H_8O_2$, for which no reliable value of E_i was found; and CS_2, left out for checking. The unbound set is formed by 14 systems having predicted negative values of ϵ_b in Table 2 of Ref. [33] (the doubtful CHF_3 is excluded). The input properties are α (isotropic), E_i, and μ, when the latter applies. As discussed in Ref. [37], in the space of descriptors the domain of each category (bound or unbound) is approximated as a region in two-dimensional (2D) space within the convex hulls of the input data separated by a dividing line. The classification of the complexes of the training set is performed as SISSO is informed of their character "bound" or "unbound" and asked to assign them to one of these two categories in an optimal manner. SISSO finds 2D descriptors yielding the maximum separation between these convex regions.

As shown in Fig. 1, the bound and unbound cases are fully separated by the descriptors $X = \frac{2\alpha+E_i}{\alpha+\mu+\mu^{1/3}}$ and $Y =$

$\frac{E_i}{\alpha} + \sqrt{E_i} + \frac{e^{E_i}}{\sqrt{E_i}}$. In general, descriptors X and Y are simple combinations of the input molecular properties. The particular analytical formulas are not relevant however, since they can change if other systems are added. Red points, representing unbound cases, are randomly scattered as expected, while blue points representing bound cases are mostly grouped in a particular region. On the other hand, test simulations not involving E_i as input property fail to separate bound and unbound systems. The equation for the dividing line is $Y = -1.333X + 5.726$, which allows checking the bound character for additional systems having properties in the range of the training set, being thus useful to experimentalists and theoreticians. The inset of Fig. 1 shows an approximate proportionality between ϵ_b and the "distance" d from each point to the dividing line, with few exceptions.

Some doubtful cases (green triangles) assigned with $\epsilon_b \geqslant 0$ in Ref. [33] are checked: C_2H_2, C_2H_6, and CH_3F. The first two are confirmed as bound in the diagram but the last is not. Also, CH_3OH, assigned with the very small $\epsilon_b = 2$ meV, lies at the bound side but very close to the dividing line. Finally, the CS_2 molecule, observed to bind e^+ in Ref. [34] and used here for checking, lies properly in the bound region in Fig. 1.

The observation of ϵ_b being a near linear function of just α for alkanes in Ref. [33], namely ($\epsilon_b = 12.4\alpha - 5.6$) meV, seems to be a consequence of E_i having almost the same value for these compounds. In fact, their general linear relation for polar and apolar molecules has a worse performance than for alkanes. The present results for general molecules seems thus to confirm the relevance of E_i, present in both descriptors, for positron binding.

All further theoretical results for polar molecules [9–22] have then been tested, with their predictions being confirmed, except for the amino acids studied in Ref. [20], due to the difficulty of obtaining reliable data for the different structures considered there.

Six additional polar molecules are tested and marked in Fig. 1. While CH_5N (methilamine, $\mu = 1.31$ D) lies close to the separation line, C_3H_7Cl (1-chloropropane, $\mu = 1.95$ D), $C_4H_{10}O$ (1-butanol, $\mu = 1.66$ D), C_3H_6S (thietane, $\mu = 1.85$ D), C_3H_8O (1-propanol, $\mu = 1.55$ D), and $C_2H_4O_2$ (acetic acid, $\mu = 1.60$ D), are clearly suggested to bind a positron, which is anticipated here. Note that only for C_3H_7Cl would binding be granted by only the dipole moment, if so.

This last point is nicely illustrated by the unbound water molecule, the only black triangle in Fig. 1. Its dipole moment of 1.85 D is larger than the critical rigid dipole of 1.65 D, while its polarizability is very small, 1.5 Å^3. The present result is the third time the complex H_2O-e^+ has been predicted as unbound, the other two being from Refs. [9] (*ab initio* quantum Monte Carlo calculation) and [33] (fitting of experimental results). The water molecule has a large ionization potential, 12.6 eV. This particular case illustrates two features simultaneously: First, the inverse proportionality between μ and α contributing to increase the value of a possibly real (nonrigid) critical dipole moment (or perhaps a narrow range for it) and, second, the importance of including E_i as a determining parameter for binding.

TABLE I. Binding energies (meV) for apolar molecules. E_i in meV and α in Å^3. Expt stands for experimental results from Refs. [33–35]; DYS refers to fitting to Eq. (5) in Ref. [33]; SG refers to Ref. [27]; TW means "this work." Molecular properties data taken from the experimental sources [33–35].

Molecules	Formula	E_i	α	Expt	DYS	SG	TW
Training set							
Butane	C_4H_{10}	10.6	8.1	35	31	26	39
Pentane	C_5H_{12}	10.4	10.0	60	54	56	54
Hexane	C_6H_{14}	10.2	11.8	80	77	87	75
Heptane	C_7H_{16}	9.9	13.7	105	100	117	105
Octane	C_8H_{18}	10.0	15.5	115	123	144	119
Nonane	C_9H_{20}	10.0	17.4	145	146	168	144
Dodecane	$C_{12}H_{26}$	9.9	22.9	220	214	222	223
Hexadecane	$C_{16}H_{34}$	9.9	30.3	310	306	264	308
Ethylene	C_2H_4	10.5	4.2	20	−17		20
Cyclopropane	C_3H_6	9.9	5.7	10	1		10
Cyclohexane	C_6H_{12}	9.9	11.1	80	68		83
Benzene	C_6H_6	9.3	10.4	150	149		148
Naphthalene	$C_{10}H_8$	8.2	16.6	300	296		301
2,3-butanedione	$C_4H_6O_2$	9.3	8.2	77	92		79
Other molecules and predictions							
Carbon disulfide	CS_2	10.1	8.7	75	98		71
Hexafluorobenzene	C_6F_6	9.9	9.8		141		84
Phosphorus chloride	Cl_3P	9.9	12.8		89		98
Cyclodecane	$C_{10}H_{20}$	10.0	18.5		160		159
Bicyclodecane	$C_{10}H_{18}$	9.4	17.7		151		215
1,3,5-hexatriene	C_6H_8	8.3	11.2		69		71
Carbon tetrabrom	CBr_4	10.3	15.3		120		123

B. Regression

Present attempts to evaluate ϵ_b with least-square ML regression for general molecules yielded a rms deviation about three times larger than the experimental accuracy, which is attributed to the presence of polar targets (see discussion below). Hence, applications are limited to apolar molecules and the training set is limited to the 14 undoubtedly bounding apolar molecules for which all properties are available [33]. Again, CS_2 [34] is left for checking. Three-dimensional regression (three descriptors) was sufficient to yield an accurate formula for ϵ_b,

$$\begin{aligned} \epsilon_b(\alpha, E_i) = {} & C_1 \mathrm{abs}\left(E_i \ln E_i - \frac{E_i \alpha}{E_i - \alpha}\right) \\ & + C_2 \mathrm{abs}\left(\frac{E_i}{\sqrt{\alpha}} - \frac{1}{E_i(\alpha - E_i)}\right) \\ & + C_3 \mathrm{abs}\left(\frac{E_i \alpha}{\alpha - E_i} - \frac{e^{E_i}}{\alpha^{1/3}}\right) + C_4, \end{aligned} \tag{1}$$

where $C_1 = 0.07706$, $C_2 = -0.96591$, $C_3 = 0.15305$, and $C_4 = 0.004247$, in a.u., and "abs" means the modulus of the expressions inside the brackets.

Equation (1) is a linear combination of additional descriptors. Its second and third terms are larger but competitive so that all terms are equally important. Again, increasing the training set would change its form, probably improving the results. Table I first shows results (last column, TW) for the training set, whose experimental (Expt) binding energies are self-consistently reproduced, with a maximum deviation of 6 meV for C_5H_{12}, within the experimental error (10 meV). For alkanes (up to hexadecane), the present results either agree within experimental error or are mostly more accurate than those (SG) recently calculated in Ref. [27]. For the other molecules of the training set, the present results are unprecedented computationally and, together with the checking system C_2S, show an impressive accordance with experimental values.

Concerning the remaining molecules in Table I, the last six predictions with formula (1) lack experimental data and are presented here as original results.

Table I also includes the results (DYS) of a linear fitting of ϵ_b in terms of α, μ (zero in this case) and N_π, namely ($\epsilon_b = 12.4\alpha + 1.6\mu + 2.4N_\pi - 5.6$) meV, in which N_π is the number of π bonds in an aromatic molecule [33]. This fitting fails for general apolar molecules, which confirms the importance of including E_i in calculations, while N_π seems probably already accounted for by combination of α and E_i effects.

Recall, however, that predictions of the present work are limited to the ranges of E_i, α and ϵ_b included in the training set. A numerical solution of Eq. (1) for $\epsilon_b = 0$ leads to a surprisingly counterintuitive accurately linear relation of the minimum value of the polarizability which allows binding for a given E_i, namely,

$$\alpha_{\min} = -2.24E_i + 27.70. \tag{2}$$

This relation, which can be also used as a bound-unbound separator within the same present ranges of properties, shows once again that binding results from a complex relation of E_i and α. In fact, the same procedure for positive ϵ_b values gives curves with hills and slopes. As a last remark, the rms deviation of our results from the experimental ones (when it applies) is, surprisingly, 3.1 meV, against the 10 meV assumed deviation for the experimental values, while the rms deviation of the DYS fitting is 13 meV. This point is suggestive that the experimental error could be even smaller.

IV. DISCUSSION AND CONCLUSIONS

The present ML-SISSO application allows useful discussion of the state of knowledge on the subject. Present methods of *ab initio* calculations for larger molecules [19,20] can treat all particles on a common footing, ascribing them orbitals and performing configuration mixing through virtual excitations. The positron then plays a singular role, occupying a more localized orbital and, concomitantly, performing as a light nucleus. This feature has a consequence on the molecular structure turning to local the polarization and electron-positron correlation effects. *Ab initio* calculations are then widely facilitated, as confirmed by the fact that they yield reliable results only for polar molecules. On the other hand, these complexes become unfriendly for modeling the interaction in terms of simple properties of isolated molecules, even for more skilled ML techniques, as noted here. Particularly, the polarization effects become more local and cannot be emulated by molecular isotropic polarizability, requiring a more sophisticated approach. Anyway, ML techniques seem appropriate to make progress along this line.

Contrarily, for apolar molecules, the positron does not localize in a particular region, visiting the whole molecule. At short distances, electron-positron correlations start being significant. These features impose huge obstacles to *ab initio* calculations, as known. But, in consequence, the isotropic α turns out to be an appropriate average measure of the attraction at medium to large distances. However, the inability to fully describe the interaction with α alone clearly demonstrates the lack of a further property for this description, connected to short-range effects, which has been shown here to be the ionization potential E_i. Furthermore, if these two quantities were fully contained to each other, SISSO would eliminate one of them. On the contrary, SISSO fails completely in cases where it is fed only the values of α. This pair of properties is necessary to emulate the physics of the interaction of e^+ with an apolar molecule.

These claims do not mean that the positron "sees" a molecule as a spherically symmetric polarizable and ionizable target. What is concluded here is that, for apolar molecules, SISSO is capable of mixing the effects of α and E_i to access the physics of the interaction, including the complicated electron-positron correlation effect, so as to yield very accurate ϵ_b values.

Equation (1) makes explicit the importance of the ionization potential E_i, which accounts here for the important short distance effects, in the evaluation of ϵ_b. This is the most important insight that comes out from the present calculations. ML-SISSO works directly with physical properties of the target, which already contains the effects of short-range repulsion and prevents the use of an unwanted free-parameter empirical cutoff potential. The physics behind the further improvements obtained here are then proposed as being a consequence of virtual Ps formation, intrinsically connected to E_i and having increasing significance as e^+ approaches the target, which defines the short-range form of the interaction.

Further investigation and applications to atoms or to enlarge the property ranges, deserve more studies. But, particularly for bound-unbound classification, ML-SISSO is shown to be already a very powerful technique. A ML-SISSO regression that includes polar molecules does not show an equivalent performance. It appears that further information concerning e^+ localization is needed, perhaps from the molecular electronic density.

Finally, beyond the present application, ML proves to be an invaluable technique not only to produce extremely accurate data but also to provide feedback for the discussion of the physics involved in the studied phenomena.

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[1] *New Directions in Antimatter Chemistry and Physics*, edited by C. Surko and F. A. Gianturco (Kluwer Academic, Dordrecht, 2001).

[2] G. F. Gribakin, J. A. Young, and C. M. Surko, Rev. Mod. Phys. **82**, 2557 (2010).

[3] G. F. Gribakin and C. M. R. Lee, Phys. Rev. Lett. **97**, 193201 (2006).

[4] T. Nishimura and F. A. Gianturco, Phys. Rev. Lett. **90**, 183201 (2003).

[5] S. d'A. Sanches, M. A. P. Lima, and M. T. do N. Varella, Phys. Rev. A **80**, 052710 (2009).

[6] O. H. Crawford, Proc. Phys. Soc., London **91**, 279 (1967).

[7] D. Assafrão and J. R. Mohallem, J. Phys. B: At., Mol. Opt. Phys. **43**, 155204 (2010).

[8] W. F. Garret, Phys. Rev. A **3**, 961 (1971).

[9] D. Bressanini, M. Mella, and G. Morosi, J. Chem. Phys. **109**, 1716 (1998).

[10] M. Mella, G. Morosi, D. Bressanini, and S. Elli, J. Chem. Phys. **113**, 6154 (2000).

[11] M. Tachikawa, R. J. Buenker, and M. Kimura, J. Chem. Phys. **119**, 5005 (2003).

[12] M. Tachikawa, R. J. Buenker, and M. Kimura, J. Chem. Phys. **121**, 9191 (2004).

[13] S. Bubin and L. Adamowicz, J. Chem. Phys. **120**, 6051 (2004).

[14] H. Chojnacki and K. Strasburger, Mol. Phys. **104**, 2273 (2006).

[15] F. A. Gianturco, J. Franz, R. J. Buenker, H.-P. Liebermann, L. Pichl, J. M. Rost, M. Tachikawa, and M. Kimura, Phys. Rev. A **73**, 022705 (2006).

[16] Y. Kita, R. Maezono, M. Tachikawa, M. Towler, and R. J. Needs, J. Chem. Phys. **131**, 134310 (2009).

[17] M. Tachikawa, Y. Kita, and R. J. Buenker, Phys. Chem. Chem. Phys. **13**, 2701 (2011).

[18] M. Tachikawa, Y. Kita, and R. J. Buenker, New J. Phys. **14**, 035004 (2012).

[19] J. Romero, J. Charry, R. Flores-Moreno, M. T. do N. Varella, and A. Reyes, J. Chem. Phys. **141**, 114103 (2014).

[20] J. Charry, J. Romero, M. T. do N. Varella, and A. Reyes, Phys. Rev. A **89**, 052709 (2014).

[21] M. Tachikawa, J. Phys.: Conf. Ser. **488**, 012053 (2014).

[22] A. S. Barbosa, S. A. Sanchez, and M. H. F. Bettega, Phys. Rev. A **96**, 062706 (2017).

[23] A. R. Swann and G. F. Gribakin, J. Chem. Phys. **149**, 244305 (2018).

[24] C. Harabati, V. A. Dzuba, and V. V. Flambaum, Phys. Rev. A **89**, 022517 (2014).

[25] X. Cheng, D. Babikov, and D. M. Schrader, Phys. Rev. A **83**, 032504 (2011).

[26] P. H. R. Amaral and J. R. Mohallem, Phys. Rev. A **86**, 042708 (2012).

[27] A. R. Swann and G. F. Gribakin, Phys. Rev. Lett. **123**, 113402 (2019).

[28] G. F. Gribakin and W. A. King, J. Phys. B: At., Mol. Opt. Phys. **27**, 2639 (1994).

[29] V. A. Dzuba, V. V. Flambaum, G. F. Gribakin, and W. A. King, Phys. Rev. A **52**, 4541 (1995).

[30] G. F. Gribakin and J. Ludlow, Phys. Rev. A **70**, 032720 (2004).

[31] J. Mitroy, M. W. J. Bromley, and G. Ryzhikh, J. Phys. B: At., Mol. Opt. Phys. **35**, R81 (2002).

[32] V. A. Dzuba, V. V. Flambaum, and G. F. Gribakin, Phys. Rev. Lett. **105**, 203401 (2010).

[33] J. R. Danielson, J. A. Young, and C. Surko, J. Phys. B: At., Mol. Opt. Phys. **42**, 235203 (2009).

[34] J. R. Danielson, J. J. Gosselin, and C. M. Surko, Phys. Rev. Lett. **104**, 233201 (2010).

[35] J. R. Danielson, A. C. L. Jones, J. J. Gosselin, M. R. Natisin, and C. M. Surko, Phys. Rev. A **85**, 022709 (2012).

[36] G. R. Schleder, A. C. M. Padilha, C. M. Acosta, M. Costa, and A. Fazzio, J. Phys. Mater. **2**, 032001 (2019).

[37] R. Ouyang, S. Curtarolo, E. Ahmetcik, M. Scheffler, and L. M. Ghiringhelli, Phys. Rev. Mater. **2**, 083802 (2018).

[38] R. Ouyang, E. Ahmetcik, C. Carbogno, M. Scheffler, and L. M. Ghiringhelli, J. Phys. Mater. **2**, 024002 (2019).

[39] http://cccbdb.nist.gov