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Temperature dependence of the complex effective piezoelectric coefficient of ferroelectric 0-3 composites

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Temperature dependence of the complex effective piezoelectric coefficient d_{31}^* for a ferroelectric 0-3 composite of small ceramic volume fraction has been studied. Theoretical predictions are based on our previously derived explicit expression of d_{31} for a dilute dispersion of spherical particles in a continuous matrix [C. K. Wong, Y. M. Poon, and F. G. Shin, Ferroelectrics **264**, 39 (2001); J. Appl. Phys. **90**, 4690 (2001)]. Comparison is made with the well-known Furukawa's model and their experimental measurements on a lead zirconate titanate (PZT)/epoxy composite with 13 vol % PZT [T. Furukawa, K. Fujino, and E. Fukada, Jpn. J. Appl. Phys. **15**, 2119 (1976)], covering a wide temperature range from −140 to +140 °C. The real part and the imaginary part of the effective piezoelectric coefficient for the composite are investigated separately. Predictions for the real part of d_{31}^* agree well with the observed values for temperatures larger than 60 °C, but are larger than the observed values for lower temperatures, while predictions for the imaginary part of d_{31}^* give fairly good agreement with the experimental data throughout the temperature range. [DOI: 10.1063/1.1503388]

I. INTRODUCTION

In previous articles,[1,2] we have derived some relatively simple explicit expressions for the effective piezoelectric d coefficients for ferroelectric 0-3 composites of small to medium-high volume fraction of the dispersed phase. The theoretical predictions have been compared with experimental results from published works, including the experimental results of Furukawa *et al.*,[3] who also presented a model for the effective piezoelectric coefficients of ferroelectric 0-3 composites. Piezoelectric composites have attracted much interest recently. They can be tailored to suit specific applications and are commonly used in sensors and actuators. The piezoelectric properties of ferroelectric 0-3 composites have been studied by many workers theoretically and experimentally.[1–9] Studies on the temperature dependence of piezoelectricity are also of great practical importance, since a sensor may operate over a wide temperature range. In addition, such studies can lead to a better understanding of these materials so that we can use them effectively in developing applications. As the temperature dependence of these effective piezoelectric coefficients for the composites involve relaxation, it is essential to consider the imaginary part of loss tangents of these coefficients. However, there are very few works concerning the imaginary part of these effective piezoelectric coefficients and their temperature dependence. Lushcheikin[10] gave the temperature dependence of the piezoelectric properties of a series of ceramic-polymer composites, but he did not consider the imaginary part of the corresponding dielectric, elastic, and piezoelectric properties. Similarly, Rittenmyer *et al.*[11] have not given the imaginary part for their measurement on piezoelectric coefficients for lead titanate/polychloroprene rubber 0-3 composites. On the other hand, Furukawa *et al.*[3] gave experimental values of the d_{31} coefficient at 50 °C for 0-3 PZT/epoxy composites of small volume fraction of inclusions, as well as the temperature profile of the complex piezoelectric d_{31}^* coefficient for the PZT/epoxy composite with 13 vol % of PZT. However, they have not compared the measured temperature dependence of the complex piezoelectric d_{31}^* coefficient with theoretical predictions based on their model.

This article examines the applicability of Furukawa's model and our model to the theoretical prediction of the temperature dependence of the effective complex d_{31}^* values for PZT/epoxy composites. Unlike our previous articles[1,2] in which real values are used for the physical properties, here complex valued properties are considered. This treatment becomes more significant for lossy materials. Experimental results from Furukawa *et al.* cited in the last paragraph are used for comparison, along with the prediction based on Furukawa's model. The experimental data were taken in a temperature range from −140 to +140 °C at 10 Hz. Both models show a fairly good agreement with the experimental data.

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However, two major differences between these two models are noted. First, the constituents are assumed to be incompressible and there is no contribution from d_{33}^{*} of the inclusion phase in Furukawa's model. In our model, the compressibility (or Poisson's ratio) of the matrix is shown to play a significant role in the real part of the composite's d_{31} constant and thus the assumption of incompressibility may limit the usage of their model for the prediction at different temperatures. Second, we have also found that the inclusion's d_{33}^{*}, which appears in our model, can have significant effects on the prediction of the imaginary part of d_{31} of the composite.

II. EFFECTIVE PIEZOELECTRIC COEFFICIENTS OF A COMPOSITE IN DILUTE LIMIT

In a previous article,[1] we have derived closed-form expressions for the effective piezoelectric coefficients d_{33}, d_{31}, and d_h for ferroelectric 0-3 composites. In our derivation, we have assumed that the inclusions are spherical, and both constituents are dielectrically and elastically isotropic. In this article, we are interested in the effective piezoelectric d_{31} coefficient only, since we are to compare the theoretical values with the complex d_{31} values of the PZT/epoxy composite measured by Furukawa *et al.*[3] In the present case, the composite contains a dilute (13 vol %) suspension of electroactive inclusions in a nonpiezoelectric matrix, thus $d_{31m}=0$ and[1]

$$d_{31}=\phi L_E\{(L_T^{\perp}+L_T^{\parallel})d_{31i}+L_T^{\perp}d_{33i}\} \tag{1}$$

where

$$L_E=\frac{3\varepsilon_m}{(1-\phi)\varepsilon_i+(2+\phi)\varepsilon_m}, \tag{2}$$

$$L_T^{\perp}=\frac{I}{1-\phi(1-3I)}-\frac{J}{1-\phi(1-3J)}, \tag{3}$$

$$L_T^{\parallel}=\frac{I}{1-\phi(1-3I)}+\frac{2J}{1-\phi(1-3J)}, \tag{4}$$

and

$$I=\frac{(1-\nu_m)Y_i}{2(1-2\nu_i)Y_m+(1+\nu_m)Y_i}, \tag{5}$$

$$J=\frac{5(1+\nu_m)(1-\nu_m)Y_i}{(1+\nu_i)(7-5\nu_m)Y_m+2(1+\nu_m)(4-5\nu_m)Y_i}, \tag{6}$$

where ε, Y, ν, and ϕ are permittivity, Young's modulus, Poisson's ratio, and volume fraction, respectively, and subscripts i and m denote inclusion and matrix, respectively.

Using an additional assumption that both phases of the composite are incompressible, Furukawa and coworkers gave an expression for the piezoelectric d coefficient[3]

$$d_{31}=\phi L_E L_T d_{31i}, \tag{7}$$

where L_E is the same as Eq. (2) and

$$L_T=\frac{5Y_i}{3Y_m+2Y_i-3\phi(Y_m-Y_i)}. \tag{8}$$

In Eq. (8), all Y's can be replaced by shear modulus.

It is noted that our expression for the effective d_{31} constant [Eq. (1)] depends not only on d_{31i}, but also d_{33i}, which is absent in Furukawa's model. In addition, our L_T's [Eqs. (3) and (4)] cannot be reduced to that of Furukawa's [Eq. (8)] by substituting $\nu_i=\nu_m=0.5$.

To investigate the temperature dependence of d_{31} based on Eqs. (1) and (7), all physical properties involved must be allowed to take on complex values. The complex permittivity, Young's modulus, Poisson's ratio, and piezoelectric coefficients may be written as [12,13]

$$\begin{cases} \varepsilon^*=\varepsilon'-i\varepsilon'' \\ Y^*=Y'+iY'' \\ \nu^*=\nu'+i\nu'' \\ d_{31}^*=d_{31}'-id_{31}'' \\ d_{33}^*=d_{33}'-id_{33}'' \end{cases}, \tag{9}$$

where the superscript asterisk indicates a complex-valued property consisting of a real part, labeled as a single-primed character, and the double-primed quantity represents its imaginary part. Suppose all quantities other than ϕ in Eqs. (1)–(6) are complex, Eq. (1) becomes

$$d_{31}^*=\phi L_E^*\{(L_T^{\perp *}+L_T^{\parallel *})d_{31i}^*+L_T^{\perp *}d_{33i}^*\} \tag{10}$$

and we define

$$\begin{cases} L_E^*\equiv L_E'+iL_E''=\dfrac{3\varepsilon_m^*}{(1-\phi)\varepsilon_i^*+(2+\phi)\varepsilon_m^*} \\ L_T^{\perp *}\equiv L_T^{\perp\prime}+iL_T^{\perp\prime\prime}=\dfrac{I^*}{1-\phi(1-3I^*)}-\dfrac{J^*}{1-\phi(1-3J^*)}, \\ L_T^{\parallel *}\equiv L_T^{\parallel\prime}+iL_T^{\parallel\prime\prime}=\dfrac{I^*}{1-\phi(1-3I^*)}+\dfrac{2J^*}{1-\phi(1-3J^*)} \end{cases} \tag{11}$$

where I^* and J^* are given by Eqs. (5) and (6), respectively, with complex-valued properties. Substituting Eqs. (11) into Eq. (10) and using Eq. (9), one can obtain

$$d_{31}'=\phi\sum_{i=1}^{8}A_i, \tag{12}$$

$$d_{31}''=\phi\sum_{i=1}^{8}B_i, \tag{13}$$

where

$$\begin{cases} A_1=L_E'(L_T^{\perp\prime}+L_T^{\parallel\prime})d_{31i}' & A_5=L_E'L_T^{\perp\prime}d_{33i}' \\ A_2=-L_E''(L_T^{\perp\prime\prime}+L_T^{\parallel\prime\prime})d_{31i}' & A_6=-L_E''L_T^{\perp\prime\prime}d_{33i}' \\ A_3=L_E'(L_T^{\perp\prime\prime}+L_T^{\parallel\prime\prime})d_{31i}'' & A_7=L_E'L_T^{\perp\prime\prime}d_{33i}'' \\ A_4=L_E''(L_T^{\perp\prime}+L_T^{\parallel\prime})d_{31i}'' & A_8=L_E''I_T^{\perp\prime}d_{33i}'' \end{cases}, \tag{14}$$

$$\begin{cases} B_1=L_E'(L_T^{\perp\prime}+L_T^{\parallel\prime})d_{31i}' & B_5=L_E'L_T^{\perp\prime}d_{33i}'' \\ B_2=L_E''(L_T^{\perp\prime\prime}+L_T^{\parallel\prime\prime})d_{31i}'' & B_6=-L_E''L_T^{\perp\prime\prime}d_{33i}'' \\ B_3=-L_E'(L_T^{\perp\prime\prime}+L_T^{\parallel\prime\prime})d_{31i}' & B_7=-L_E'L_T^{\perp\prime\prime}d_{33i}' \\ B_4=-L_E''(L_T^{\perp\prime}+L_T^{\parallel\prime})d_{31i}' & B_8=-L_E''L_T^{\perp\prime}d_{33i}' \end{cases}. \tag{15}$$

Similarly for Furukawa's model, Eqs. (7) and (8) become

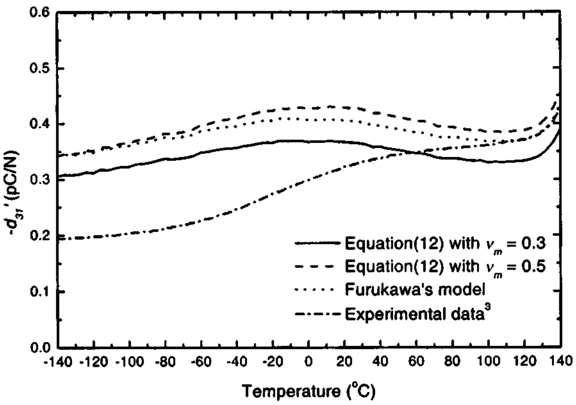

FIG. 1. Comparison with experimental data of Furukawa *et al.*[3] for the effective d_{31} (real part) of a PZT/epoxy composite measured at 10 Hz.

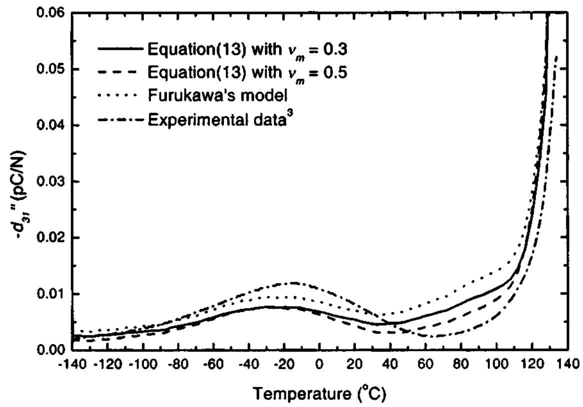

FIG. 2. Comparison with experimental data of Furukawa *et al.*[3] for the effective d_{31} (imaginary part) of a PZT/epoxy composite measured at 10 Hz.

$$d_{31i}^* = \phi L_E^* L_T^* d_{33i}^*, \tag{16}$$

$$L_T^* \equiv L_T' + iL_T'' = \frac{5Y_i^*}{3Y_m^* + 2Y_i^* - 3\phi(Y_m^* - Y_i^*)}, \tag{17}$$

and d_{31}' and d_{31}'' for Furukawa's model are

$$d_{31}' = \phi \sum_{i=1}^{4} C_i, \tag{18}$$

$$d_{31}'' = \phi \sum_{i=1}^{4} D_i, \tag{19}$$

where

$$\begin{cases} C_1 = L_E' L_T' d_{31i}' & C_3 = L_E' L_T'' d_{31i}'' \\ C_2 = -L_E'' L_T'' d_{31i}' & C_4 = L_E'' L_T' d_{31i}'' \end{cases}, \tag{20}$$

$$\begin{cases} D_1 = L_E' L_T' d_{31i}'' & D_3 = -L_E' L_T'' d_{31i}' \\ D_2 = -L_E'' L_T'' d_{31i}'' & D_4 = -L_E'' L_T' d_{31i}' \end{cases}. \tag{21}$$

In summary, Eqs. (12) and (13) are used for the prediction of effective piezoelectric coefficients d_{31}' and d_{31}'', with L_E^* and L_T^*'s given by Eqs. (11). For Furukawa's model, Eqs. (18) and (19) are to used accordingly for d_{31}' and d_{31}'', with L_E^* and L_T^* given by Eqs. (11) and (17), respectively.

III. COMPARISON WITH EXPERIMENTAL DATA

Our theoretical prediction and the prediction based on Furukawa's model of complex piezoelectric coefficient $d_{31}^* = d_{31}' - id_{31}''$ are compared with the experimental data of Furukawa *et al.*[3] for a PZT/epoxy composite for the temperature range between -140 and $+140$ °C (Figs. 1 and 2), with frequency at 10 Hz. The composite sample has 13% of PZT by volume. As insufficient data on dielectric/elastic/piezoelectric properties of the constituent materials (PZT and epoxy) were provided in their articles, typical values have been adopted in our calculation, as will be explained in the following paragraphs (Secs. III A and III B).

A. Temperature dependence of complex ε_i and d_{33i}

Temperature dependence of the permittivity ε_i^* of PZT before poling was given in the article of Furukawa *et al.*,[3] but only two sets of after-poling values (corresponding to two temperature values) were given. We assume that the ratio of the before-poling value to the after-poling value is uniform throughout the temperature range, and is the same for the real and the imaginary parts. This ratio is taken to be the average of the two ratios (1.256 and 1.295) associated with the two temperatures given by Furukawa *et al.* Thus, a factor of 1.28 is multiplied to the before-poling values of ε_i in the computation.

Concerning the piezoelectric coefficient d_{33i}^*, no experimental results have been reported by Furukawa *et al.* and the results reported by other researchers must be used. It should be noted that, for a given temperature, the dielectric, elastic, and piezoelectric properties vary with the composition of PZT,[14,15] as well as the addition of dopants.[16,17] Zhang *et al.*[16] have measured the ε_i, d_{31i}, and d_{33i} for four types of PZT from 4.2–300 K. The magnitude and profile of their experimental ε_i and d_{31i} for their Navy-type II PZT are very similar to the corresponding results of PZT given by Furukawa *et al.*[3] Thus, in this work the percentage changes of d_{33i}' from -140 °C to room temperature are assumed to be the same as the results of Zhang *et al.* of d_{33}. For d_{33i}' at a higher-temperature range, the percentage changes of d_{33} measured from room temperature to 100 °C for PZT 802 given by Cheng[18] is used, and the whole temperature profile of d_{33i}' is scaled in such a way that $d_{33i}' = 400$ pC/N at 50 °C, a value we have used before.[1,2] For temperatures greater than 100 °C, the variation of d_{33i}' with temperature is assumed to be the same as that of d_{31i}'.

Concerning the d_{33i}'', although there have been some experimental works on the temperature dependence of d_{33} and d_{31} for PZT,[3,14–22] very few have reported the loss tangent or the imaginary part of d_{33} for PZT. These experimental results reveal that the temperature profile of d_{33i}' is quite similar to that of d_{31i}'. We assume the temperature profile of the loss tangent of d_{33i} to be similar to that of d_{31i}, which has been measured by Furukawa *et al.*[3] The whole temperature profile of d_{33i}'' is then scaled in such a way that the loss tangent at room temperature agrees with the measurement of Wang *et al.*[23] for PZT 400. Wang *et al.* have reported the d_{33} values and their loss tangents for PZT 400 and PZT 5H in their article. The d_{33} values are 253 and 590 pC/N for PZT 400

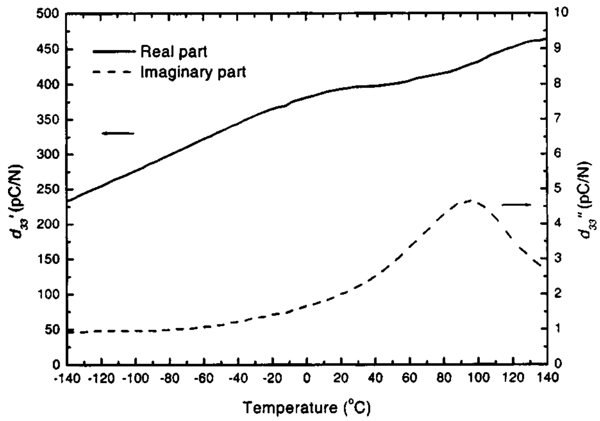

FIG. 3. Temperature dependence of piezoelectric coefficient d_{33} for PZT ceramic.

and PZT 5H, respectively. We use the loss tangent for PZT 400 (rather than that of PZT 5H) because its room temperature d_{33} value is comparable to the corresponding value (221.5 pC/N) measured by Cheng[18] for PZT 802. Based on the above assumptions, the temperature profile of the complex d_{33i} is shown in Fig. 3.

B. Temperature dependence of mechanical properties of constituents

Furukawa *et al.*[3] have measured the temperature dependence of the complex Young's modulus for PZT and epoxy, but they have not measured the Poisson's ratios. Poisson's ratio for the PZT ν_i has been assumed to be 0.3,[1,2] and its variation with temperature is assumed to be small. Moreover, as the Poisson's ratio for the epoxy ν_m may vary drastically with temperature, two extreme values, 0.3 and 0.5, have been plotted in Figs. 1 and 2 for comparison. We note that Furukawa *et al.* have used $\nu_i = \nu_m = 0.5$, regardless of the temperature variation. Nevertheless, Fig. 1 shows that the band enclosed by our predictions may not be narrow enough to confirm that the Poisson's ratio for the constituents are temperature independent. According to the measurement of Tcharkhtchi *et al.*[24] for epoxy (which is also diglycidylether of bisphenol A, as Furukawa *et al.*), the bulk modulus is free of viscoelastic effects. From their results of bulk modulus and the measurements of Furukawa *et al.* of Young's modulus, we find that the imaginary part of the Poisson's ratio for the epoxy is a very small value (about 0.002) and it does not affect the d'_{31} and d''_{31} values of the composite significantly. Concerning the imaginary part of the Poisson's ratio for the PZT, we expect that its effect on the prediction of the effective d^*_{31} should be smaller than that of epoxy. It is because the complex Young's modulus of the PZT has been shown to be nearly independent of temperature,[3] and the present composite sample contains quite a small volume fraction of PZT. Therefore, we have assumed the imaginary part of the Poisson's ratio for the constituents to be zero, which is the same as the treatment used in Menard's text.[25] It should be noted that some types of PZT may not behave like the sample from Furukawa *et al.* (negligible temperature dependence for Young's modulus). In such a case, the above treatment might not be appropriate.

C. Temperature dependence of effective d_{31} constants

From Fig. 1, concerning the real part of d_{31}, the prediction based on Furukawa's model lies in between our predictions for $\nu_m = 0.3$ and $\nu_m = 0.5$. At the low-temperature regime, ν_m tends to be smaller and therefore its actual value should be closer to 0.3 rather than 0.5. At the high-temperature regime, especially near 140 °C, where a primary dispersion is seen in the Young's modulus for epoxy,[3] ν_m tends to 0.5. Our previous articles[1,2] have reported that our model is slightly closer to the experimental data (measured at 50 °C) than Furukawa's model, with $\nu_m = 0.35$. For the region from room temperature to about 110 °C, which is the glass transition temperature for the epoxy,[26] variation of ν_m is expected to be small and our predictions are in relatively good agreement with the experimental data. For the low-temperature region, both Furukawa's model and our theory do not make a good agreement with the experimental data. Furukawa *et al.* have suggested that the predictions were greater than the experimental values due to imperfect poling of the inclusions.[3] This argument does not seem to be applicable in this system because we have shown that theoretical predictions give good agreement at some temperature ranges, but they disagree with experimental data at some other temperature ranges. An imperfect poling should not influence only some temperature ranges (say, around −100 and 50 °C that Furukawa *et al.*[3] reported); it should influence the whole range from −140 to 140 °C. However, we believe that there are likely other mechanisms not included in the present model which may have been significant at low temperatures, as the discrepancy between predictions and the experimental data tends to be larger there (Fig. 1). This may need further investigation.

On the other hand, Fig. 1 also reveals the importance of ν_m in d'_{31} prediction. As ν_m is expected to vary with temperature, the actual prediction varies across the band enclosed by $\nu_m = 0.3$ and $\nu_m = 0.5$ and it is quite wide when compared to that in Fig. 2. Both real and imaginary parts of our L_T's vary with ν_m, with $L_T^{\perp\prime}$ showing the largest variation. We have found that $L_T^{\perp\prime}$ and $L_T^{\perp\prime\prime}$ will increase in magnitude by about 250%–225% and 93%–91%, respectively, for ν_m changing from 0.3–0.5. However, $L_T^{\parallel\prime}$ and $L_T^{\parallel\prime\prime}$ change at most by 0.7% and 15%, respectively. Figure 4 shows the relative contributions of the various terms (only A_1, A_5, and C_1 are shown; other A's and C's are very small) defined in Eqs. (14) and (20) to the resultant d'_{31}. It clearly reveals that A_5 varies quite substantially with ν_m. No factor similar to A_5 appears in the model of Furukawa *et al.* since it is associated with d_{33i} which is not in their model. Actually, for the assumption of rigid inclusions ($Y_i \gg Y_m$), which should be applicable to Furukawa's experimental data, Eqs. (5) and (6) may be approximated to

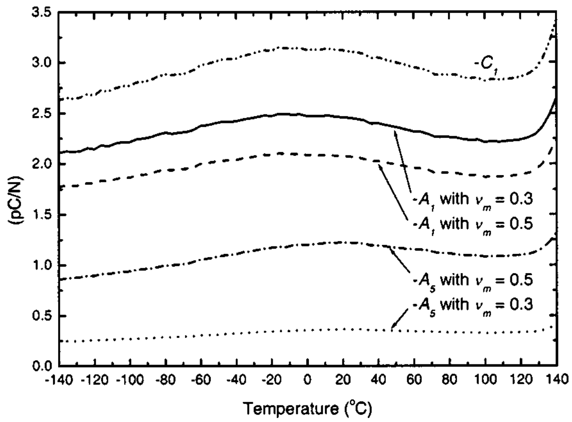

FIG. 4. Prediction for the temperature dependence of A_1, A_5, and C_1 [Eqs. (14) and (20)]. Other A's and C's are not shown because they contribute to less than 0.1% to the overall values.

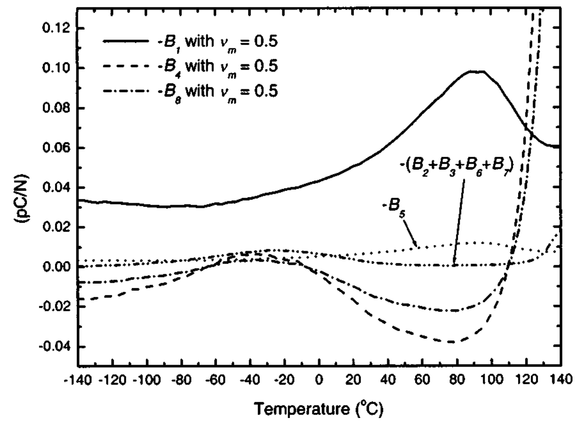

FIG. 6. Prediction for the temperature dependence of B's [Eq. (15)] with $\nu_m = 0.5$.

$$I \approx \frac{1-\nu_m}{1+\nu_m}, \tag{22}$$

$$J \approx \frac{5}{2}\,\frac{1-\nu_m}{4-5\nu_m}, \tag{23}$$

without dependence on all other mechanical properties except ν_m. It shows that ν_m is a significant parameter in such predictions of piezoelectric coefficients of 0–3 composites. Furukawa's model assumes the constituents are incompressible. Their model may then be thought to be applicable only to high temperatures where ν_m is close to 0.5. Indeed, the predictions given by Furukawa's model are in excellent agreement with the experimental data at temperatures beyond 110 °C (the glass transition temperature for the epoxy).

Figure 2 shows the imaginary part of d_{31} for the composite, comparing Furukawa's model and our prediction. The profile of the experimental result looks very similar to the imaginary part of the permittivity of epoxy shown in the article by Furukawa *et al.*[3] Below room temperature, both Furukawa's model and our prediction give fairly good agreement to the experimental data. Concerning the temperature range from 50 to about 120 °C, our prediction based on $\nu_m = 0.5$ allows a direct comparison with Furukawa's model. As evident from Fig. 2, our predictions show significant advantage over Furukawa's model. The predicted values assuming $\nu_m = 0.5$ are much closer to the experimental data than the predicted values for $\nu_m = 0.3$ in this temperature range. A feature in our expression [Eq. (10)] is that, other than d^*_{31i}, d^*_{33i} plays an important role in the prediction, especially in the imaginary part. Figure 5 and 6 show the relative contributions of the various terms defined in Eqs. (15) to the resultant d''_{31} for $\nu_m = 0.3$ and 0.5, respectively. Compared with Fig. 7, which shows relative contributions of the terms in Eqs. (21) from Furukawa's model, they clearly reveal that B_5 and B_8 of Eqs. (15) vary sensitively with ν_m (though the effective d''_{33} may not be so), and they are both factors associated with d_{33i} which is not included in Furukawa's model. This d_{33i} contribution is more significant in the higher-temperature region.

All in all, the temperature profile of the complex d_{31} constant for the composite is quite similar to the temperature profile of complex permittivity for the epoxy as shown in the article by Furukawa *et al.*[3] However, the drastic variation of the composite d^*_{31} should not be mostly governed by the piezoelectric properties of PZT alone, but also the dielectric and elastic properties of the constituents. At such small volume fraction of PZT, the dielectric and elastic properties of

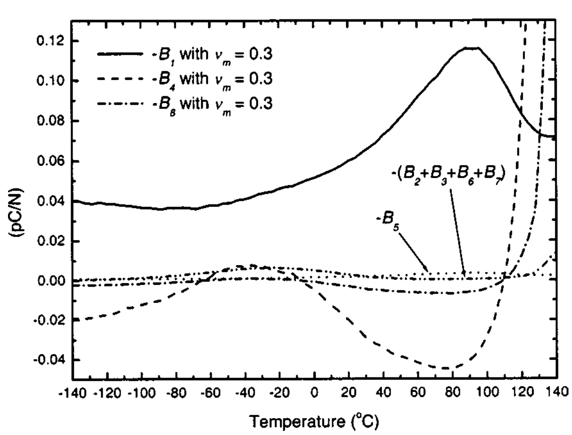

FIG. 5. Prediction for the temperature dependence of B's [Eq. (15)] with $\nu_m = 0.3$.

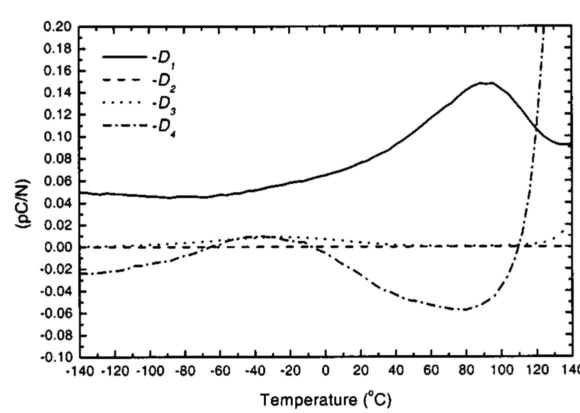

FIG. 7. Prediction for the temperature dependence of D_1 to D_4 [Eq. (21)].

the matrix phase are thought to make larger contributions to the composite d_{31}^* than that of PZT's. Our predictions give slightly better agreement with experimental data over a broader temperature range. This merit is much more obvious in the imaginary part of the effective piezoelectric coefficient d_{31}.

IV. CONCLUSIONS

The temperature dependence of the complex effective piezoelectric coefficient d_{31}^* for ferroelectric 0–3 composites has been studied theoretically at small ceramic volume fraction. Our theoretical prediction has been compared with Furukawa's model for the experimental values of PZT/epoxy from published work, also by Furukawa *et al.* Fairly good agreement was obtained for temperatures larger than 60 °C for the prediction of the real part as well as the imaginary part of d_{31}^*. The comparison shows that the contribution of the d_{33}^* term in Eq. (10) (which is absent in Furukawa's model) can be significant in predicting the imaginary part of the effective d_{31} coefficient. Moreover, the compressibility of the matrix is shown to be a key factor for the real-part prediction of the d_{31} constant, especially for composites with rigid inclusions. As incompressibility for both constituents is not assumed in our model, our predictions are expected to be applicable to a wider temperature range. To conclude, temperature affects the piezoelectric properties of a composite in a complicated manner because the piezoelectric properties of a composite depend not only on the piezoelectric properties of the constituents, but also on their dielectric and elastic properties, although they contribute to varying degrees.

ACKNOWLEDGMENT

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